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[Molecules] Manuscript ID: molecules-1368447 - Submission Received

Editorial Office <molecules@mdpi.com> Fri, Aug 20, 2021 at 2:55 PM
Reply-To: molecules@mdpi.com
To: Diding Suhandy <diding.sughandy@fp.unila.ac.id>
Cc: Meinilwita Yulia <meinilwitayulia@polinela.ac.id>

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Thank you very much for uploading the following manuscript to the MDPI submission system. One of our editors will be in touch with you soon.

Journal name: Molecules
Manuscript ID: molecules-1368447
Type of manuscript: Article
Title: Quantification of Corn Adulteration in Wet and Dry Processed Peaberry Ground Roasted Coffees by UV Spectroscopy and Chemometrics
Authors: Meinilwita Yulia, Diding Suhandy *
Received: 20 August 2021
E-mails: meinilwitayulia@polinela.ac.id, diding.sughandy@fp.unila.ac.id
Submitted to section: Analytical Chemistry,
https://www.mdpi.com/journal/molecules/sections/Analytical_Chemistry
5th Anniversary of Analytical Chemistry Section—Recent Advances in Analytical Chemistry
https://www.mdpi.com/journal/molecules/special_issues/5th_Analytical_Chemistry

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[Molecules] Manuscript ID: molecules-1368447 - Assistant Editor Assigned

Gabriela Ledwójcik <ledwojcik@mdpi.com> Fri, Aug 20, 2021 at 4:56 PM
Reply-To: ledwojcik@mdpi.com
To: Diding Suhandy <diding.sughandy@fp.unila.ac.id>
Cc: Gabriela Ledwójcik <ledwojcik@mdpi.com>, Meinilwita Yulia <meinilwitayulia@polinela.ac.id>, Molecules Editorial Office <molecules@mdpi.com>

Dear Dr. Suhandy,

Your manuscript has been assigned to Gabriela Ledwójcik for further processing who will act as a point of contact for any questions related to your paper.

Journal: Molecules
Manuscript ID: molecules-1368447
Title: Quantification of Corn Adulteration in Wet and Dry Processed Peaberry Ground Roasted Coffees by UV Spectroscopy and Chemometrics
Authors: Meinilwita Yulia, Diding Suhandy *

Received: 20 August 2021
E-mails: meinilwitayulia@polinela.ac.id, diding.sughandy@fp.unila.ac.id

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Assistant Editor
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[Molecules] Manuscript ID: molecules-1368447 - Major Revisions

Molecules Editorial Office <molecules@mdpi.com> Fri, Sep 10, 2021 at 7:51 PM
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To: Diding Suhandy <diding.sughandy@fp.unila.ac.id>
Cc: Meinilwita Yulia <meinilwitayulia@polinela.ac.id>, Molecules Editorial Office <molecules@mdpi.com>

Dear Dr. Suhandy,

Thank you again for your manuscript submission:

Manuscript ID: molecules-1368447
Type of manuscript: Article
Title: Quantification of Corn Adulteration in Wet and Dry Processed Peaberry Ground Roasted Coffees by UV Spectroscopy and Chemometrics
Authors: Meinilwita Yulia, Diding Suhandy *
Received: 20 August 2021
E-mails: meinilwitayulia@polinela.ac.id, diding.sughandy@fp.unila.ac.id
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Cc: Meinilwita Yulia <meinilwitayulia@polinela.ac.id>

Dear Dr. Suhandy,

Thank you very much for resubmitting the modified version of the following manuscript:

Manuscript ID: molecules-1368447
Type of manuscript: Article
Title: Quantification of Corn Adulteration in Wet and Dry Processed Peaberry Ground Roasted Coffees by UV Spectroscopy and Chemometrics
Authors: Meinilwita Yulia, Diding Suhandy *
Received: 20 August 2021
E-mails: meinilwitayulia@polinela.ac.id, diding.sughandy@fp.unila.ac.id
Submitted to section: Analytical Chemistry,
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Cc: Meinilwita Yulia <meinilwitayulia@polinela.ac.id>, Molecules Editorial Office <molecules@mdpi.com>

Dear Dr. Suhandy,

Thank you very much for providing the revised version of your paper:

Manuscript ID: molecules-1368447
Type of manuscript: Article
Title: Quantification of Corn Adulteration in Wet and Dry Processed Peaberry Ground Roasted Coffees by UV Spectroscopy and Chemometrics
Authors: Meinilwita Yulia, Diding Suhandy *
Received: 20 August 2021
E-mails: meinilwitayulia@polinela.ac.id, diding.sughandy@fp.unila.ac.id
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Cc: Molecules Editorial Office <molecules@mdpi.com>

Dear Dr. Suhandy,

Thank you very much for submitting your manuscript to Molecules:

Journal name: Molecules
Manuscript ID: molecules-1368447
Type of manuscript: Article
Title: Quantification of Corn Adulteration in Wet and Dry Processed Peaberry Ground Roasted Coffees by UV Spectroscopy and Chemometrics
Authors: Meinilwita Yulia, Diding Suhandy *
Received: 20 August 2021
E-mails: meinilwitayulia@polinela.ac.id, diding.sughandy@fp.unila.ac.id
Submitted to section: Analytical Chemistry,
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[Molecules] Manuscript ID: molecules-1368447 - Funding Information Confirmation

Molecules Editorial Office <molecules@mdpi.com> Wed, Oct 6, 2021 at 1:18 PM
Reply-To: molecules@mdpi.com, ledwojcik@mdpi.com
To: Meinilwita Yulia <meinilwitayulia@polinela.ac.id>, Diding Suhandy <diding.sughandy@fp.unila.ac.id>
Cc: Molecules Editorial Office <molecules@mdpi.com>, Gabriela Ledwójcik <ledwojcik@mdpi.com>

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Funding information in our system:
the Indonesian Ministry of Education, Culture, Research, and Technology (KEMENDIK-BUDRISTEK): Grant No. 378.1/PL15.8/PT/2021

Manuscript ID: molecules-1368447
Type of manuscript: Article
Title: Quantification of Corn Adulteration in Wet and Dry Processed Peaberry Ground Roasted Coffees by UV Spectroscopy and Chemometrics
Authors: Meinilwita Yulia, Diding Suhandy *
Received: 20 August 2021
E-mails: meinilwitayulia@polinela.ac.id, diding.sughandy@fp.unila.ac.id
Submitted to section: Analytical Chemistry,
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[Molecules] Manuscript ID: molecules-1368447 - Final Proofreading Before Publication

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Cc: Meinilwita Yulia <meinilwitayulia@polinela.ac.id>, Molecules Editorial Office <molecules@mdpi.com>

Dear Dr. Suhandy,

We invite you to proofread your manuscript to ensure that this is the final version that can be published and confirm that you will require no further changes from hereon:

Manuscript ID: molecules-1368447
Type of manuscript: Article
Title: Quantification of Corn Adulteration in Wet and Dry-Processed Peaberry Ground Roasted Coffees by UV–Vis Spectroscopy and Chemometrics
Authors: Meinilwita Yulia, Diding Suhandy *
Received: 20 August 2021
E-mails: meinilwitayulia@polinela.ac.id, diding.sughandy@fp.unila.ac.id
Submitted to section: Analytical Chemistry,
https://www.mdpi.com/journal/molecules/sections/Analytical_Chemistry
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Kind regards,

Ms. Gabriela Ledwójcik, M.Sc.
Assistant Editor
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MDPI Branch Office, Kraków
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[Molecules] Manuscript ID: molecules-1368447 - Accepted for Publication

Molecules Editorial Office <molecules@mdpi.com> Wed, Oct 6, 2021 at 1:18 PM
Reply-To: Gabriela Ledwójcik <ledwojcik@mdpi.com>, Molecules Editorial Office <molecules@mdpi.com>
To: Diding Suhandy <diding.sughandy@fp.unila.ac.id>
Cc: Meinilwita Yulia <meinilwitayulia@polinela.ac.id>, Molecules Editorial Office <molecules@mdpi.com>, Gabriela Ledwójcik <ledwojcik@mdpi.com>

Dear Dr. Suhandy,

Congratulations on the acceptance of your manuscript, and thank you for your interest in submitting your work to Molecules:

Manuscript ID: molecules-1368447
Type of manuscript: Article
Title: Quantification of Corn Adulteration in Wet and Dry Processed Peaberry Ground Roasted Coffees by UV Spectroscopy and Chemometrics
Authors: Meinilwita Yulia, Diding Suhandy *
Received: 20 August 2021
E-mails: meinilwitayulia@polinela.ac.id, diding.sughandy@fp.unila.ac.id
Submitted to section: Analytical Chemistry,
https://www.mdpi.com/journal/molecules/sections/Analytical_Chemistry
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[Molecules] Manuscript ID: molecules-1368447 - Manuscript Resubmitted

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To: Diding Suhandy <diding.sughandy@fp.unila.ac.id>
Cc: Meinilwita Yulia <meinilwitayulia@polinela.ac.id>

Dear Dr. Suhandy,

Thank you very much for resubmitting the modified version of the following manuscript:

Manuscript ID: molecules-1368447
Type of manuscript: Article
Title: Quantification of Corn Adulteration in Wet and Dry-Processed Peaberry Ground Roasted Coffees by UV–Vis Spectroscopy and Chemometrics
Authors: Meinilwita Yulia, Diding Suhandy *
Received: 20 August 2021
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Submitted to section: Analytical Chemistry,
https://www.mdpi.com/journal/molecules/sections/Analytical_Chemistry
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Manuscript ID: molecules-1368447
Type of manuscript: Article
Title: Quantification of Corn Adulteration in Wet and Dry Processed Peaberry Ground Roasted Coffees by UV Spectroscopy and Chemometrics
Authors: Meinilwita Yulia, Diding Suhandy *
Received: 20 August 2021
E-mails: meinilwitayulia@polinela.ac.id, diding.sughandy@fp.unila.ac.id
Submitted to section: Analytical Chemistry,
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Dear Authors,

We are pleased to inform you that your article "Quantification of Corn Adulteration in Wet and Dry-Processed Peaberry Ground Roasted Coffees by UV–Vis Spectroscopy and Chemometrics" has been published in Molecules as part of the Special Issue 5th Anniversary of Analytical Chemistry Section—Recent Advances in Analytical Chemistry and is available online:

Abstract: https://www.mdpi.com/1420-3049/26/20/6091
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Kind regards,

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Journal: Molecules (https://www.mdpi.com/journal/molecules) (ISSN 1420-3049)

Manuscript ID: molecules-1368447

Type: Article

Title: Quantification of Corn Adulteration in Wet and Dry Processed Peaberry Ground Roasted Coffees by UV Spectroscopy and Chemometrics

Authors: Meinilwita Yulia , Diding Suhandy *

Abstract: In this present research, a spectroscopic method based on UV spectroscopy was utilized to quantify the level of corn adulteration in peaberry ground roasted coffee by chemometrics. Peaberry coffee with two types of bean processing of wet and dry processed methods was used and intentionally adulterated by corn with 10-50% level of adulteration. UV spectral data was obtained for aqueous samples in the range between 250 and 400 nm with a 1 nm interval. Three multivariate regression methods including partial least squares regression (PLSR), multiple linear regression (MLR), and principal component regression (PCR) were used to predict the level of corn adulteration. The result showed that all individual regression models using individual wet and dry samples are better than that of global regression models using combined wet and dry samples. The best calibration model for individual wet and dry and combined samples were obtained for the PLSR model with a coefficient of determination more than 0.70 and RMSE below 6% (w/w) for both calibration and validation. However, the error prediction in terms of RMSEP and bias were highly increased when the individual regression model was used to predict the level of corn adulteration with differences in the bean processing method. The obtained results demonstrated that the use of the global PLSR model is better in predicting the level of corn adulteration. The error prediction for this global model was acceptable with low RMSEP and bias for both individual and combined prediction samples. The obtained RPDp and RERp in prediction for the global PLSR model was more than 2 and 5 for individual and combined samples, respectively. The proposed method using UV spectroscopy with a global PLSR model can be applied to quantify the level of corn adulteration in peaberry ground roasted coffee with different bean processing methods.

Author's Reply to the Review Report (Reviewer 1)

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	Yes	Can be improved	Must be improved	Not applicable
Does the introduction provide sufficient background and include all relevant references?	(x)	( )	( )	( )
Is the research design appropriate?	(x)	( )	( )	( )
Are the methods adequately described?	(x)	( )	( )	( )
Are the results clearly presented?	(x)	( )	( )	( )
Are the conclusions supported by the results?	(x)	( )	( )	( )

Comments and Suggestions for Authors

The article is very interesting and well-written. Here are some minor comments:

1. I have some concerns about using SG first derivative to correct for baseline. This removes only constant trend. Is it the case for your UV-VIS spectra. There are many better methods suitable for baseline removal, no matter what function it follows. For instance methods with asymmetric penalised least squares usually perform better. Have you tried any of these?
2. line140: Should be "spectral" instead of "spectra"

3. Line 142: Which of the variables were then finally chosen for PCR? How high loadings were considered, i.e. what threshold was the cutoff?
4. Line 207: Should be Figure 3a
5. How were the data prepared before PCA apart from preprocessing methods?
6. line 213: Should be Figure 3b
7. Line 215: Should be Figure 4
8. Line 251: Should be Table 3
9. Line 292: Should be Table 4

Submission Date	20 August 2021
Date of this review	10 Sep 2021 14:02:00

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Author's Reply to the Review Report (Reviewer 2)

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Does the introduction provide sufficient background and include all relevant references?	(x)	( )	( )	( )
Is the research design appropriate?	( )	( )	(x)	( )
Are the methods adequately described?	(x)	( )	( )	( )
Are the results clearly presented?	( )	( )	(x)	( )
Are the conclusions supported by the results?	( )	( )	(x)	( )

Comments and Suggestions for Authors

This manuscript reported corn adulteration in peaberry coffees by UV-Vis spectroscopy. This work is helpful to establish a reliable, low-cost method in coffee adulteration method. However, the overall scientific value of this work remains incomplete. Some of the issues should be resolved.

This work achieved quantification as their primary method towards adulteration. However, classification using PCA scores and PLS-DA should be evaluated when detecting fraud. Based on Figure 5 and Table 4, the overall PLSR for quantification still yields high

errors in the test sets, which may be the shortcoming as UV-Vis is low-cost and imprecise. However, again, it should be justified the actual classification between pure/adulterated coffee at the claimed LOD around 20% is significant. Otherwise, this work provides few useful information for practical application.

Although this work focused on UV frequency range, most UV spectroscopy also extends to the Visible region. Additionally, the visual appearance for adulteration is different enough in own observation. The author should refer UV as UV-Vis all along the manuscript. Also, the full UV-vis range should be inspected for modelling, at least solid reasons should be given why the visible range is not considered, along with the data.

Submission Date	20 August 2021
Date of this review	25 Aug 2021 05:43:56

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Author's Reply to the Review Report (Reviewer 3)

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* Author's

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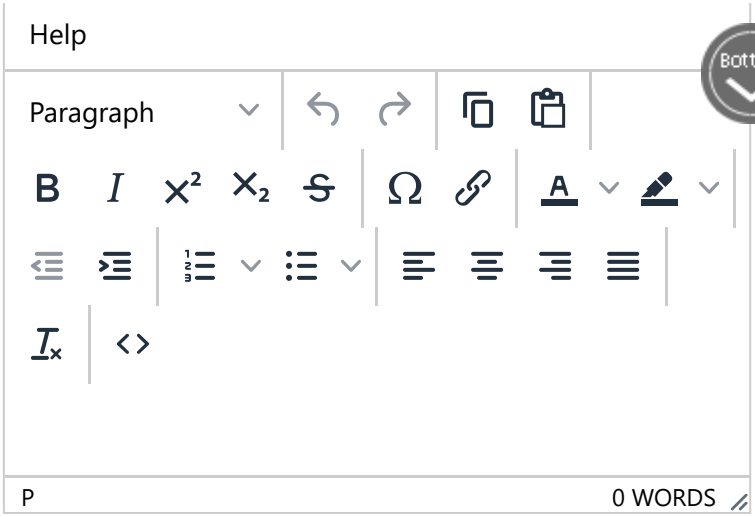

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	Yes	Can be improved	Must be improved	Not applicable
Does the introduction provide sufficient background and include all relevant references?	( )	(x)	( )	( )
Is the research design appropriate?	( )	( )	(x)	( )
Are the methods adequately described?	( )	(x)	( )	( )
Are the results clearly presented?	(x)	( )	( )	( )
Are the conclusions supported by the results?	( )	(x)	( )	( )

Comments and Suggestions for Authors

The present manuscript presents a method for the quantification of corn adulteration in peaberry ground roasted coffee that have been wet or dry processed. The work uses multivariate linear regression methods to model and predict the amount of adulteration, and PLS has proved to be the best method for both wet and dry processed samples.

The manuscript is very well written and is very clear throughout the text. However, even though the premise is very interesting (to use a more affordable approach, such as UV spectroscopy, for the detection of the adulteration), it carries some flaws that I believe are critical to the acceptance of the results. Hence, I do not recommend its publication in the current form. My points will be presented in the document attached.

peer-review-14086141.v1.pdf
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Submission Date	20 August 2021
Date of this review	31 Aug 2021 01:10:57

The present manuscript presents a method for the quantification of corn adulteration in peaberry ground roasted coffee that have been wet or dry processed. The work uses multivariate linear regression methods to model and predict the amount of adulteration, and PLS has proved to be the best method for both wet and dry processed samples.

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i) Major flaws in interpreting the spectra and the loadings information throughout the text. As described in section 2.3., the data was pre-processed by smoothing, standardization, baseline correction and differentiation. The last method is, indeed, much used for regression. The first derivative of a spectrum will show how it varies, where the peaks are presented as zero (since it is the local maxima), the regions where the original spectrum signal are increasing are presented as positive values and the regions where the original spectrum signal are decreasing are presented as negative values. Figure 2(a) and 2(b) clearly shows that the pre-processed data presents the first derivative. The "peaks" in the pre-processed spectra (270, 290, 315 and 345 nm) are not peaks in the raw spectra. Interpretations were made in sections 3.1. and 3.2., where these values were treated, discussed, and compared to the literature. This should be closely looked by the authors, so no misinterpretation remains. If comparisons should be made, it should take into consideration that the data represents the first derivative.

ii) Why was PCA only conducted with the adulterated samples? Was it important to separate the wet from dry sample or to separate the pure from adulterated sample? I believe that the latter is the most important one. As I will discuss in the next point, calibration was not good because the variability between each replicate sample was very high. Take Figure 5 as an example. For solutions with the same amount of adulteration, the model did not see them as equal, but as quite different. Perhaps lack of reproduction is related to the variability of the beans? Is it related to any of the process conducted? The fact is that the solutions that should represent the same thing does not. That is what the regression error is so big. Not because it is not good, or invalid, but because it is trying to model data that are not good. Perhaps the objective of the work shouldn't be to quantify (because it is not predicting anything, since the LOQ is even higher than the range studied), but to classify the samples using multivariate methods. If the "pure" sample data were added to the PCA, maybe PCs will be able to separate them from the adulterated ones. This could be a tool to detect if it is the real peaberry coffee beans or not. PCA could even be refined to try to separate the samples in groups (e.g., based on the amount of corn bean).

iii) The regression is not quantifying anything! As I mentioned in the point above, it is very difficult to accept a model in which the LOQ is substantially higher than the maximum concentration value used for the construction of the model. Your consideration that it needs a future improvement would be reasonable, but only if your own model could be validated. The results show that you would only be able to quantify adulterations over 70%. However, you never used any solutions above this concentration. Your model for quantification was constructed entirely below the limit that it says that it can quantify. When you compare your model to other methods in the literature

that present such small LOD, it seems that, even though the UV spectroscopy is more accessible, it is not worth to substitute using another technique such as NMR or LIBS.

Minor points for authors to review:

i) The abstract says that R^2 for PLSR is “more than 0.70”, but in fact is way more than that.

ii) Introduction can be improved to give a background to the reader regarding the corn beans as adulterants. You only cite that “the adulteration is frequently happened in the form of ground roasted coffee”. How important is the adulteration with corn beans? Is it relevant?

iii) The method is not clear regarding how you achieve 100 and 99 samples, when you describe that it was prepared only the 10%, 20%, 30%, 40% and 50% concentrations. The reader can only guess that all are replicates when looking at Figure 5. Why were this procedure chosen? Why not use other concentrations?

iv) Page 3, line 124 says that 3 pre-processing methods were used, but 4 are described.

v) Page 4, lines 148 and 149 says that three samples were selected for the calibration set out of 5. From what is show in Table 1, this is probably mistaken. I believe it should say that 4 out of 5 was chosen as the calibration set.

vi) Page 8, lines 270-282 are unnecessary in my opinion. Comparing the behavior of adulterations in honey, measured by ion mobility, shouldn’t be the same as in the system studied in this work.

vii) It is not clear to what variables were used for the MLR model. Which frequency was chosen for that? Information should be provided.

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Is the research design appropriate?	(x)	( )	( )	( )
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Are the results clearly presented?	(x)	( )	( )	( )
Are the conclusions supported by the results?	(x)	( )	( )	( )

Comments and Suggestions for Authors	The questions and concerns are clearly responded to and revised during this revision, thus this manuscript can be published.
Submission Date	20 August 2021
Date of this review	22 Sep 2021 13:32:27

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Article

Quantification of Corn Adulteration in Wet and Dry Processed Peaberry Ground Roasted Coffees by UV-Vis Spectroscopy and Chemometrics

Meinilwita Yulia [1] and Diding Suhandy [2,*]

[1] Department of Agricultural Technology, Lampung State Polytechnic, Jl. Soekarno Hatta No. 10, Rajabasa Bandar Lampung 35141, Indonesia; meinilwitayulia@polinela.ac.id
[2] Department of Agricultural Engineering, Faculty of Agriculture, The University of Lampung, Jl. Soemantri Brojonegoro No.1, Bandar Lampung 35145, Indonesia; diding.sughandy@fp.unila.ac.id
* Correspondence: diding.sughandy@fp.unila.ac.id; Tel.: +62-0813-7334-7128

Abstract: In this present research, a spectroscopic method based on UV-Vis spectroscopy was utilized to quantify the level of corn adulteration in peaberry ground roasted coffee by chemometrics. Peaberry coffee with two types of bean processing of wet and dry processed methods was used and intentionally adulterated by corn with 10-50% level of adulteration. UV-Vis spectral data was obtained for aqueous samples in the range between 250 and 400 nm with a 1 nm interval. Three multivariate regression methods including partial least squares regression (PLSR), multiple linear regression (MLR), and principal component regression (PCR) were used to predict the level of corn adulteration. The result showed that all individual regression models using individual wet and dry samples are better than that of global regression models using combined wet and dry samples. The best calibration model for individual wet and dry and combined samples were obtained for the PLSR model with a coefficient of determination in the range of ~~more than~~ 0.83~0.93~~70~~ and RMSE below 6% (w/w) for ~~both~~ calibration and validation. However, the error prediction in terms of RMSEP and bias were highly increased when the individual regression model was used to predict the level of corn adulteration with differences in the bean processing method. The obtained results demonstrated that the use of the global PLSR model is better in predicting the level of corn adulteration. The error prediction for this global model was acceptable with low RMSEP and bias for both individual and combined prediction samples. The obtained RPD_P and RER_P in prediction for the global PLSR model was more than 2 and 5 for individual and combined samples, respectively. The proposed method using UV-Vis spectroscopy with a global PLSR model can be applied to quantify the level of corn adulteration in peaberry ground roasted coffee with different bean processing methods.

Keywords: UV-Vis spectroscopy; peaberry coffee; individual model; global model; dry bean processing; wet bean processing; adulteration; authentication; partial least squares regression; multiple linear regression

Citation: Yulia, M; Suhandy, D. Title. *Molecules* **2021**, *26*, x. https://doi.org/10.3390/xxxxx

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Received: date
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1. Introduction

Specialty coffee is a premium product and according to the Specialty Coffee Association of Europe [1], "Specialty coffee is defined as a crafted quality coffee-based beverage, which is judged by the consumer (in a limited marketplace at a given time) to have a unique quality, a distinct taste and personality different from, and superior to, the common coffee beverages offered. The beverage is based on beans that have been grown in an accurately defined area, and which meet the highest standards for green coffee and its roasting, storage, and brewing." In Indonesia, specialty coffee can be *Coffea liberica*, *Coffea arabica,* or *Coffea canephora*. In the market, three types of commercially traded specialty

coffee are available: single-origin coffees (including Gayo coffee, Kalosi coffee, Mandailing coffee, Toraja coffee, Lampung coffee), digested animal coffees (including wild civet coffee, feeding civet coffee, bat coffee) and peaberry coffee (a single bean/monocotyledon) [2–3].

Nowadays, the growth of specialty coffee consumption is faster than that of the traditional one [2]. Mostly driven by economic motivation, food fraud both in terms of mislabeling and adulteration is now increasing and becoming a serious problem in specialty coffee trading. For example, it was reported that 42% of commercial civet coffee was fake or adulterated with normal non-civet coffee [4]. For peaberry specialty coffee, the adulteration is frequently happened in the form of ground roasted coffee, since after roasting and grinding, the discrimination of ground coffee made from peaberry and traditional (normal) coffee is almost impossible with the conventional methods [5–6]. For this reason, several sensitive emerging analytical methods to quantify adulterants in coffee have been developed in the past ten years: high-performance liquid chromatography (HPLC) [7], gas chromatography-mass spectrometry (GC-MS) [8], electrospray ionization mass spectrometry (ESI-MS) [9], and real-time polymerase chain reaction (RT-PCR) [10]. However, these accurate methods are expensive in the instrumentation and required a highly trained person.

Spectroscopic based method using different electromagnetic regions along with chemometrics has been successfully applied for cereal adulteration quantification in ground roasted coffee both in single and multiple adulterants using near-infrared (NIR), ultraviolet-visible (UV-Vis), mid-infrared, Raman, nuclear magnetic resonance (NMR), and laser-induced breakdown spectroscopy (LIBS) [11–16]. Most of these methods are less expensive in the device and faster in sample preparation (little or no need for sample preparation). Some previous works have incorporated the variation of postharvest treatments in coffee samples such as differences in coffee roasting (light, medium, and dark) in the developed calibration model [17–18]. However, in the aforementioned studies, no reported works included the influence of other important postharvest factors especially the bean processing method in the developed calibration models. Previously, Suhandy and Yulia [19] showed a significant influence of differences in bean processing method (dry, wet, and semi-dry) on the discrimination of Lampung robusta specialty ground roasted coffee. For green bean coffee, Barrios-Rodríguez et al. [20] successfully demonstrated the significant discrimination between the wet, dry and semi-dry of *Coffea arabica L.* var. Colombia using infrared spectroscopy coupled with chemometrics.

In this study, corn was selected as an adulterant material due to its low cost and huge availability in the Indonesian market. Additionally, corn is one of the most used diluents in coffee adulteration as reported in several previous works [15-16, 21-23]. Therefore, in this present research, we evaluated a spectroscopic method based on UV-Vis spectroscopy and chemometrics to quantify the corn adulteration in coffee involving two common types of bean processing of wet and dry-processed methods. In more specific, the objective of this study is to investigate a robust calibration model using three different linear regression methods including PLSR, MLR, and PCR for quantification of the corn adulteration in peaberry specialty coffee incorporated with different ~~in~~ bean processing methods.

2. Materials and Methods

2.1. Peaberry samples and their adulteration

Green bean peaberry coffee samples with two types of bean processing method (wet and dry with about 1 kg each) were obtained from a certified coffee supplier located in Garut, West Java province, Indonesia. The peaberry green bean samples are specialty grade from mixed cultivars of *Coffea arabica L.* and its hybrid (mostly Sigarar utang, Lini S, Ateng super, Catimor, and Typica) harvested in the year 2019 and originated from Cikuray, Papandayan, and Kamojang mountainous coffee plantation in Garut, West Java

province, Indonesia (latitude and longitude coordinates 7°19'22.4"S and 107°51'37.9"E, respectively; altitude, ± 1,600 m).

Before roasting by portable roaster (at 200°C for 15 minutes), all beans were visually inspected and showed no defective grains. After roasting, the unroasted and over-roasted beans were removed carefully by hand. After grinding, particles of size 40 mesh (400 µm) were obtained, which were used to perform all physicochemical analyzes.

Corn with its low cost and huge availability in the Indonesian market was selected as an adulterant. Corn samples were collected from a local farmer in Lampung province, Indonesia. According to Sezer et al. [16] with modification, corn was roasted in two steps: at 100°C for 7 minutes and followed by 200°C for 10 minutes, ground (Sayota home grinder) and mechanically sieved through a U.S. mesh size 40 to obtain the same particle size for all the samples (400 µm). The wet and dry processed peaberry ground roasted coffees were intentionally adulterated with the ground roasted corn in the range of 10-50% (w/w) with an increment of 10% (w/w). This adulteration range was chosen according to several previous works [15–16]. It is also the most common adulteration level found in the Indonesian markets [12].

Total 199 samples (1 gram each) of adulterated peaberry dry and wet-processed coffees were provided. It consists of 20 samples for each level of corn adulteration resulted in a total of 100 samples for dry-processed peaberry coffees and 99 samples for wet-processed peaberry coffees (19 samples were provided at a level of 40% for wet-processed peaberry coffees). ~~Total 100 and 99 samples (1 gram each) of adulterated peaberry wet and dry processed coffees were provided.~~ Figure 1 shows its visual appearance with 10-50% of corn adulteration level before extraction with hot distilled water. The adulterated peaberry wet-processed samples are darker than that of peaberry dry ones. However, it is visually difficult to discriminate between the different levels of adulteration in both wet and dry adulterated peaberry samples.

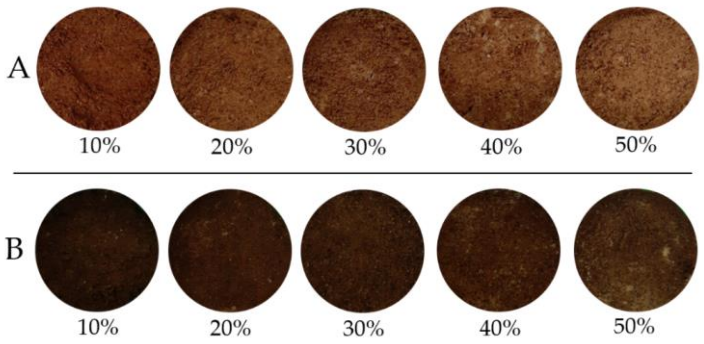


Figure 1. The visual appearance of peaberry wet-processed (A) and dry-processed (B) coffee with 10-50% of corn adulteration.

2.2. Sample extraction and UV-Vis spectral data measurement

Coffee samples were extracted based on a standard procedure as reported by previous studies [5, 12]. Raw UV-Vis spectral data was obtained for aqueous samples in the range between 250 and 400 nm with 1 nm interval using a UV-visible spectrometer (Genesys™ 10S UV-Vis, Thermo Scientific, Waltham, MA, USA) in .csv format. After reformatting into .xls, the raw spectral data were imported to the Unscrambler X ver. 10.4 (CAMO Software AS, Oslo, Viken, Norway) for chemometrics analysis.

2.3. Chemometrics

Since there is no standard protocol for spectral pre-processing, a trial and error approach ~~were~~ was adopted. Different spectral pre-preprocessing is available in the Unscrambler X ver. 10.4 (CAMO Software AS, Oslo, Viken, Norway) to reduce or to remove the effect of several different unwanted interfering phenomena such as particle size influence (baseline different and light scattering), etc. As mentioned by Roger et al. [24] and Bian et al. [25], it is hard to determine which pre-processing can successfully improve the given original spectral data. For this reason, instead of selecting the best pre-processing, to optimize the effect of spectral pre-processing, the combination of several spectral pre-processing was often used [19]. To eliminate noise and systematic spectra variation, three consecutive spectral pre-processing were found to be the best applied: moving averaging smoothing with 5 segments (MAS), standard normal variate (SNV), and Savitzky-Golay first derivative with 11 smoothing gaps~~,~~ and second-order polynomial (SG1d). MAS was widely used to smooth the spectral data before applying various pre-processing [2~~6~~1]. SNV was effective to normalize spectra for canceling the scattering effect while SG1d was used to correct the baseline effect [2~~7~~2–2~~8~~3]. Due to similarity in coffee species of both samples of wet and dry-processed coffees, it was expected that the spectral difference in peaberry coffee samples due to differences in the level of adulteration between wet and dry-processed coffees was small. The SG1d spectral pre-processing was also used to enhance these small spectral differences [26].

PCA (principal component analysis), which is widely used in analytical chemistry [18], was used to study any possible clustering of adulterated peaberry samples according to the differences in bean processing methods. The plot of the score and its corresponding x-loadings from the first two principal components (PCs) was presented for raw and pre-processed spectra.

Among numerous multivariate linear regression methods for quantification of adulteration in coffee, the partial least squares regression (PLSR) is widely used. In this research, we apply PLSR and compare it to other linear methods of multiple linear regression (MLR) and principal component regression (PCR) to quantify the level of corn adulteration. PLSR and PCR were developed using spectral data from 250 to 400 nm (number of variables=161). In MLR, a selected few variables were obtained from a plot of x-loadings. Wavelengths that are associated with the positive and negative peaks ~~(both positive and negative) with high x-loadings~~ were used as input. All regression models were validated by the full cross-validation method to optimize the model parameters.

According to Costa et al. [2~~9~~4] and Macedo et al. [30~~25~~], the samples were manually selected and separated into two sets: calibration and prediction set as presented in Table 1. The procedure of this separation of the samples was as follows: order the samples concerning the corn adulteration level (from minimum to maximum values), then four~~three~~ samples were selected every five samples for the calibration and the rest for prediction. By doing this, as seen in Table 1, a more uniform of the calibration and prediction sample sets could be obtained.

Table 2 shows the statistical parameters used to assess the quality of the calibration model and evaluate the performance of its prediction [31~~26~~–32~~27~~]. For model evaluation the following statistical parameters were used including the coefficient of determination of calibration and cross-validation (R^2_c and R^2_{cv}), root means squared errors of calibration, and cross-validation (RMSEC and RMSECV), and the ratio of prediction to deviation in cross-validation (RPD_{cv}). Limit of detection (LOD) and limit of quantification (LOQ) were also calculated according to Milani et al. [15] and Rambla-Alegre et al. [33].

In the prediction step, the performance of the regression model was evaluated using the following statistical parameters: the coefficient of determination for prediction (R^2_P), standard error of prediction (SEP), bias, root mean square error of prediction (RMSEP), RPD and RER in prediction. The RPD is the ratio of the standard deviation of reference data for the validation or prediction set to RMSECV or RMSEP and the RER is the ratio between the difference of the maximum and minimum reference values for the data in the

prediction set to RMSEP [3428]. ~~Limit of detection (LOD) and limit of quantification (LOQ) were also calculated according to Milani et al. [15] and Rambla-Alegre et al. [29].~~

Table 1. Individual and global peaberry wet and dry processed samples with 10-50% of corn adulteration in calibration and prediction sets. The range, mean and standard deviation were expressed in % (w/w).

Individual wet samples	**Calibration set**	**Prediction set**
Number of samples	83	16
Range	10-50	10-50
Mean	29.88	30.00
Standard deviation (SD)	14.36	14.14
Individual dry samples		
Number of samples	84	16
Range	10-50	10-50
Mean	30.00	30.00
Standard deviation (SD)	14.31	14.14
Global samples		
Number of samples	167	32
Range	10-50	10-50
Mean	29.94	30.00
Standard deviation (SD)	14.29	13.91

Table 2. Statistical parameters and their equations are used to assess the calibration model and its prediction performance.

Steps	**Parameters**	**Equations** [1]	**Accepted values**
Calibration	R^2_c and R^2_{cv}	$1-\frac{\sum_{i=1}^{n}(y_i-\hat{y}_i)^2}{\sum_{i=1}^{n}(y_i-\bar{y}_i)^2}$	Close to 1
	RMSEC and RMSECV	$\sqrt{\frac{\sum_{i=1}^{n}(y_i-\hat{y}_i)^2}{n}}$	As low as possible
	RPD_{cv}	$\frac{SD_{cv}}{RMSCV}$	More than 2
	LOD	$\frac{3\sigma}{S}$	As low as possible
	LOQ	$\frac{10\sigma}{S}$	As low as possible
Prediction	RMSEP	$\sqrt{\frac{\sum_{i=1}^{n}(y_i-\hat{y}_i)^2}{n}}$	As low as possible
	SEP	$\sqrt{(RMSEP)^2-(bias)^2}$	As low as possible
	bias	$(\check{y}-\bar{y})$	Close to 0
	RPD_p	$\frac{SD_{pred}}{RMSEP}$	More than 2
	~~RER_p~~	~~$\frac{y_{max}-y_{min}}{RMSEP}$~~	~~More than 10~~
	~~LOD~~	~~$\frac{3\sigma}{S}$~~	~~As low as possible~~
	RER_p~~LOQ~~	$\frac{y_{max}-y_{min}}{RMSEP}$ ~~$\frac{10\sigma}{S}$~~	More than 10~~As low as possible~~

[1] n : number of samples
y_i : actual corn adulteration values
$\hat{y}_i$: predicted corn adulteration values
$\check{y}_i$: mean of predicted corn adulteration values

$\bar{y}_i$: mean of actual corn adulteration values
σ : standard deviation of residual between actual and predicted corn adulteration values or SECP
S : the slope of the regression line

2.4. Software

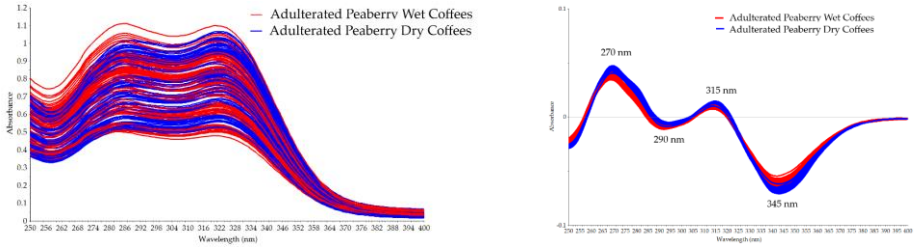
Chemometrics and spectral analysis were calculated using the Unscrambler X ver. 10.4 (CAMO Software AS, Oslo, Viken, Norway).

3. Results and Discussion

3.1. Spectral data of wet and dry peaberry coffees with different levels of corn adulteration

Figure 2 (a) shows the typical raw spectral data of adulterated peaberry wet and dry coffees in the range between 250-400 nm. Our spectra were similar to the work reported by Souto et al. [350]. The raw spectra are broad and overlap, hence it is hard to differentiate between wet and dry adulterated peaberry. A better visualization was obtained using the preprocessed spectra as seen in Figure 2 (b). In general, the intensity of absorbance in dry adulterated peaberry coffees was higher than that of the wet one and it is in line with the previously reported work [19].

Several positive and negative peaks were observed clearly in the pre-processed spectral data (MAS+SNV+SG1d). The highest positive peak at 270 nm of pre-processed spectra was closely related to the C=O chromophore in caffeine molecules as reported by some previous works [350–361], indicating the significant difference of the caffeine content in adulterated wet and dry peaberry coffees. The negative peaks at 290 and 345 nm of pre-processed spectra was corresponding with the absorbance of chlorogenic acids– (CGA) of raw UV-Vis spectra in previous work– [361]. Navarra et al. [372] reported a wavelength at 330 nm for the CGA absorbance when ethanol was used as the solvent. Dankowska et al. [361] reported wavelength at 320 nm as one of the negative peaks found in the raw UV-Vis spectral data of arabica and robusta coffee and its adulteration using water as solvent. In this study, with water used as the solvent, the peak of CGA of pre-processed spectral data was shifted to the longer wavelength at 345 nm. This shifting phenomenon was ~~supported~~ also found by the previous work by Souto et al. [350], with water used as the solvent, they found wavelength shifting of CGA from 320 nm to 325 nm in raw UV-Vis spectral data.



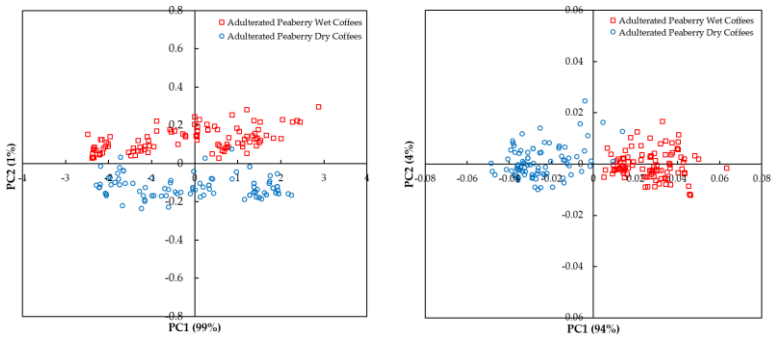
(**a**) (**b**)

Figure 2. Spectral data of peaberry wet and dry-processed coffee with 10-50% of corn adulteration in the range between 250 and 400 nm: (**a**) Raw spectra; (**b**) Pre-processed spectra (MAS+SNV+SG1d).

3.2. PCA scores and loadings

Figure 32 (a) shows the scores of the first two principal components (PC1 and PC2) of all coffee samples including wet and dry with 10-50% of corn adulteration using raw spectral data in the range between 250 and 400 nm. Explained variance for the PC1 was

obtained high (PC1=99%). However, in the term of PC1, good separation between the adulterated peaberry wet and dry coffees could not be achieved. A better PCA score plot was achieved using pre-processed spectral data in the range between 250 and 400 nm as presented in Figure 3~~2~~ (b). Along the PC1 with 94% explained variance, all of the adulterated peaberry wet samples were plotted to the right of PC1 (PC1 positive). While most of the adulterated peaberry dry samples were on the left of PC1 (PC1 negative). Figure ~~3~~4 shows the loadings plot of PC1 and PC2 using pre-processed spectral data. This plot shows the contribution of PC1 and PC2 to the separation of the adulterated peaberry wet and dry samples. In PC1 and PC2, the positive peaks with positive loading were observed at a wavelength of 267 and 345 nm. These wavelengths could be related to the absorbance of chlorogenic acids and trigonelline content in arabica coffee (CGA) [3~~5~~0], indicating that the adulterated peaberry wet samples coffees contain high contents of these compounds. This result was supported by previously reported work. Comparing to the semi-dry method, Duarte et al. [3~~3~~8] reported that the wet coffees processed method showed higher contents of CGA and trigonelline due to loss of other components with higher water solubility by lixiviation and thermal degradation during the wet processing. Three peaks with negative loadings were observed at wavelengths of 278, 290, and 328 nm. These wavelengths mainly contributed to discriminate against the adulterated peaberry dry coffees. Souto et al. [3~~5~~0] reported the maxima electronic absorption of trigonelline at 275 nm, caffeine at 280 nm, and caffeic acid at 325 nm using raw UV-Vis spectra. However, the adulterated peaberry dry coffees were mainly discriminated by the negative peak for PC1 at the wavelength of 278 nm, indicating that the adulterated peaberry dry samples coffees contain high contents of caffeine. It was supported by previous work [3~~9~~4]. It was reported that the caffeine content in dry processing coffees is higher since about 40% of caffeine is removed with pulp during the wet processing [3~~9~~4]. These positive and negative peaks obtained from PCA x-loadings of pre-processed spectral data at 267, 278, 290, 305, 328, and 345 nm were used as input variables for constructing the MLR model.



(**a**) (**b**)

Figure 3. Plot of the first two principal components by PCA in the range between 250 and 400 nm: (**a**) Raw spectra; (**b**) Pre-processed spectra (MAS+SNV+SG1d).

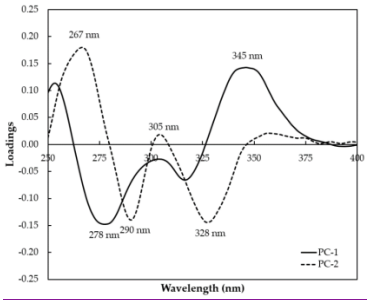
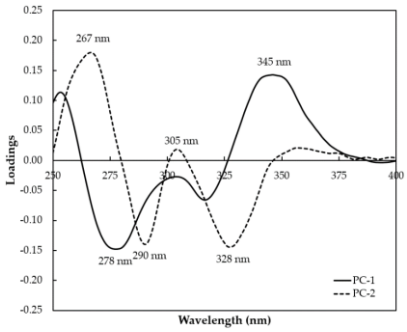


Figure 4. The plot of x-loading calculated by PCA in the range between 250 and 400 nm using pre-processed spectra.



~~**Figure 4.** The plot of x-loading calculated by PCA in the range between 250 and 400 nm using pre-processed spectra.~~

3.3. Model development for quantification of corn adulteration

The correlation between pre-processed UV-Vis spectral data and level of corn adulteration was quantified by developing three types of multivariate regression including PLSR, MLR, and PCR using calibration sample set and validated with full-cross validation method. Three types of models were developed according to the range of samples: individual wet model, individual dry model, and global model. For individual wet and dry models, the multivariate regression was developed using individual wet (n=83) and dry (n=84) calibration samples, respectively. For the global model, the multivariate regression was developed using a combined sample of wet and dry calibration samples (n=167). The results are presented in Table 3~~2~~. In general, all developed regression models had a sufficient and acceptable number of latent variables (LVs) ranging from 4 to 9. It meets with the number of LVs not exceed 15 as indicated by Bureau et al. [40~~35~~]. A small difference between the RMSEC and RMSECV was also observed for the PLSR and PCR model indicating the optimal number of LVs could be obtained [40~~35~~]. The best individual wet model was obtained for the PLSR model using five LVs (explained 98% of the accumulated variance of the spectrum data and 92% of the score data) with R^2_c=0.93, R^2_{cv}=0.89, RMSEC=3.85% (w/w), and RMSECV= 4.80% (w/w). For dry samples, the best individual dry model was also obtained for the PLSR model using six LVs (explained 97% of the accumulated variance of the spectrum data and 94% of the score data) with less accuracy than the individual wet model. However, both individual wet and dry models are acceptable with RPD higher than 2, indicating that PLS-DA models can be classified as

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excellent [41~~36~~]. In a previous study, Sezer et al. [16] reported a similar result for quantification of *Coffee arabica* adulteration with corn samples employing laser-induced breakdown spectroscopy (LIBS) and PLS regression with R^2_{cal}=0.995, R^2_{val}=0.990, RMSEC=4.32% (w/w), and RMSECV= 4.84% (w/w) could be obtained. A better result was shown by Winkler-Moser et al. [17] for predicting corn adulteration using NIR spectroscopy with lower error both in calibration and validation. They obtained PLS model with R^2_{cal}=0.979, R^2_{val}=0.974, RMSEC=1.05% (w/w), and RMSECV=1.17% (w/w).

It is noted that compared to the global model, all developed individual regression models using individual wet and dry samples are better in accuracy with higher R^2 and lower error (both in terms of RMSEC and RMSECV). ~~A similar phenomenon was reported in a previous study. Aliano-Gonzalez et al. [37] reported quantification of adulteration in honey using ion mobility and the PLS method with five types of adulterants.~~ The PLS model was created to quantify both individual adulterant and global (combined) adulterants. The result showed that the PLS model for the quantification of individual adulterants was better than that of combined adulterants. According to Table 2, the PLSR model was superior compared to other regression models for the individual wet, dry and global regression model. The RPD in cross-validation was more than 2 in all PLSR regression models (RPD critical = 2.0 [31-~~4~~32]). According to Kapper et al. [42~~38~~], all developed PLSR models showed good accuracy with a high coefficient of determination between actual and predicted corn adulteration ($R^2 \geq 0.70$) both in calibration and validation.

Table 3. Model development results for adulteration quantification using partial least square regression (PLSR), multiple linear regression (MLR), and principal component regression (PCR) with the individual and combined~~global~~ sample set using pre-processed spectra (MAS+SNV+SG1d). The best model for each regression method is highlighted in bold. The RMSEC and RMSECV were expressed in % (w/w).

Model	Regression Method	LVs	R^2_c	R^2_{cv}	RMSEC	RMSECV	RPD_{cv}
Individual wet model	**PLSR**	**5**	**0.93**	**0.89**	**3.85**	**4.80**	**2.99**
	MLR		0.87	0.87	5.44	5.20	2.76
	PCR	8	0.90	0.87	4.57	5.17	2.78
Individual dry model	**PLSR**	**6**	**0.92**	**0.89**	**3.93**	**4.87**	**2.94**
	MLR		0.84	0.84	6.00	5.75	2.49
	PCR	9	0.90	0.88	4.46	5.05	2.83
Global model	**PLSR**	**8**	**0.88**	**0.83**	**4.93**	**5.86**	**2.44**
	MLR		0.63	0.63	8.87	8.68	1.65
	PCR	9	0.72	0.69	7.52	8.02	1.78

Figure 5 shows plots of the best PLSR calibration model for individual wet, dry, and combined calibration samples. Visually, it can be noticed that the residuals of calibration were randomly scattered closely to the regression line (bias is close to 0) for individual and combined calibration samples. The SEC and slope for individual calibration wet samples were 3.87% (w/w) and 0.93 resulting in the LOD and LOQ of 12.48% (w/w) and 41.61% (w/w), respectively. Similarly, the LOD and LOQ for individual calibration dry samples were 12.88% (w/w) and 42.93% (w/w). For combined calibration samples, the LOD and LOQ were 16.84% (w/w) and 56.14% (w/w). Comparing to previous works, our result was inferior. For example, Milani et al. [15] reported satisfactory LOD values of 0.31-0.86% using the NMR spectroscopy with a different roasting profile. Sezer eta l. [16] reported a quantitative approach using LIBS for coffee adulteration with different adulterants (corn, wheat, and chickpea) and resulted in a promising result with the LOD below 0.6% could be obtained. The obtained LOD and LOQ using UV-Vis spectroscopy in this study were in the range of 12.48%~16.84% and 41.61%~56.14%. In this present study, a calibration and validation regression model was developed using corn adulterated samples in the range of 10-50% (w/w). However, the obtained LOD and LOQ in this study suggested we extend the range of corn adulteration up to more than 50%. For this reason,

it needs an improvement for practical application. However, in Indonesia, adulteration more than 50% of specialty coffee is commonly found for economically motivated adulteration [12].

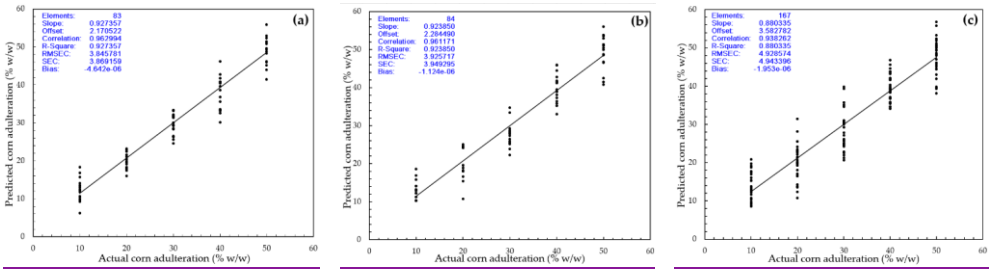


Figure 5. Actual versus predicted values of corn adulteration (% w/w) in peaberry coffee samples for the best PLSR model on (a) individual wet calibration samples (b) individual dry calibration samples (c) combined calibration samples.

3.4. Prediction using individual and global PLSR models

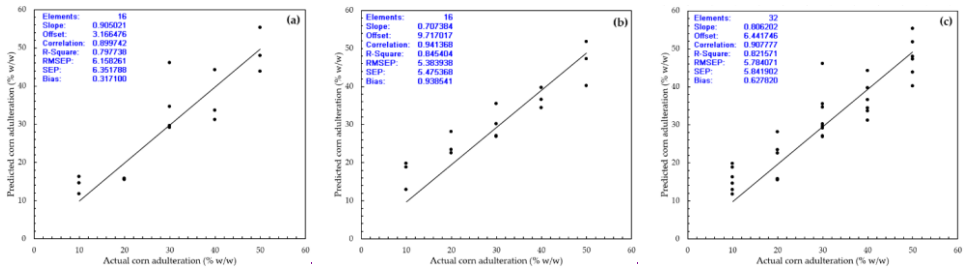
To evaluate the influence of bean processing on the performance of the developed calibration model in prediction corn adulteration, the prediction was calculated on the individual (n=16 for both individual wet and dry samples) and combined prediction sample set (n=32). The best individual and global PLSR models were used as input. The results are presented in Table 4~~3~~. The individual wet PLSR model resulted in high RPD (RPD_P=3.96) when it was used to predict corn adulteration in individual wet samples. However, this model failed to predict corn adulteration in individual dry samples resulting in low RPD_P (the RPD is less than 1). The error prediction in terms of RMSEP and bias was highly increased. A similar result was found for prediction using the individual dry PLSR model. The individual dry PLSR model showed a good prediction with RPD_P=3.33 for prediction of dry samples and a failed prediction with RPD_P=0.28 for prediction of wet samples. The use of the global PLSR model is promising. The error prediction for this global model was acceptable with low RMSEP and bias for both individual and combined prediction samples. The RPD_P was higher than 2 for both individual predictions of wet and dry samples and combined samples. According to Chang et al. [43] and Valinger et al. [44] models with RPD >2.0 are excellent descriptions and predictions of experimental data. In terms of RER, the global PLSR with RER in the range of 3 to 10 for both individual and combined prediction samples, which is classified as a good practical utility model according to Jia et al. [45~~39~~].

Table 4. Prediction results for individual and combined prediction samples using the best individual and global PLSR models. The SEP, RMSEP, and bias were expressed in % (w/w).

Individual wet PLSR model	**SEP**	**RMSEP**	**Bias**	**RPD_P**	**RER_P**
Wet prediction samples	3.64	3.57	0.56	3.96	11.20
Dry prediction samples	11.43	45.59	-44.22	0.31	0.88
Combined prediction samples	24.23	32.33	-21.83	0.43	1.24
Individual dry PLSR model	**SEP**	**RMSEP**	**Bias**	**RPD_P**	**RER_P**
Wet prediction samples	9.61	50.96	50.10	0.28	0.78
Dry prediction samples	4.36	4.24	0.36	3.33	9.43
Combined prediction samples	26.31	36.16	25.23	0.38	1.11
Global PLSR model	**SEP**	**RMSEP**	**Bias**	**RPD_P**	**RER_P**
Wet prediction samples	6.35	6.16	0.32	2.30	6.49
Dry prediction samples	5.48	5.38	0.94	2.63	7.43

Combined prediction samples	5.84	5.78	0.63	2.41	6.92

~~Figure 5 shows plots of prediction for individual and combined prediction samples predicted using the global PLSR model. Visually, it can be noticed that the residuals of prediction were randomly scattered closely to the regression line (bias is close to 0) for individual and combined prediction samples. The SEP and slope for individual prediction wet samples were 6.35% (w/w) and 0.91 resulting in the LOD and LOQ of 20.93% (w/w) and 69.78% (w/w), respectively. Similarly, the LOD and LOQ for individual prediction dry samples were 23.15% (w/w) and 77.18% (w/w). For combined prediction samples, the LOD and LOQ were 21.63% (w/w) and 72.10% (w/w). Comparing to previous works, our result was inferior. For example, Milani et al. [15] reported satisfactory LOD values of 0.31–0.86% using the NMR spectroscopy with a different roasting profile. Sezer eta l. [16] reported a quantitative approach using LIBS for coffee adulteration with different adulterants (corn, wheat, and chickpea) and resulted in a promising result with the LOD below 0.6% could be obtained. The obtained LOD and LOQ using UV spectroscopy in this study in the range of 20.93%~23.15% and 69.78%~72.10% need an improvement for practical application. However, in Indonesia, adulteration more than 50% of specialty coffee is commonly found for economically motivated adulteration [12].~~



~~**Figure 5.** Actual versus predicted values of corn adulteration (% w/w) in peaberry coffee samples using FPLSR global model on (a) individual wet prediction samples (b) individual dry prediction samples (c) combined prediction samples.~~

4. Conclusions

This research describes the use of UV-Vis spectroscopy along with chemometrics to quantify the level of corn adulteration in peaberry specialty coffee with different bean processing methods. The proposed UV-Vis spectroscopy and global PLSR model detected an admixture of corn in the peaberry ground roasted coffee in the range 10% to 50% with the LOD values of 12.48%~16.84% ~~20.93–23.15%~~ could be reported for individual and combined samples. The reliability of the global PLSR model was confirmed by external validation using both individual (wet and dry) and combined prediction samples indicating the great potential of UV-Vis spectroscopy and chemometrics as a green and low-cost analytical method for authentication of peaberry specialty ground roasted coffee incorporated with different in bean processing method.

Author Contributions: Conceptualization, D.S., and M.Y.; methodology, D.S.; software, M.Y.; validation, M.Y.; formal analysis, M.Y., and D.S.; investigation, M.Y.; resources, D.S.; data curation, M.Y.; writing—original draft preparation, D.S., and M.Y.; writing—review and editing, D.S., and M.Y.; visualization, M.Y., and D.S.; supervision, D.S.; project administration, D.S.; funding acquisition, M.Y. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

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Response to Reviewer 1 Comments

The article is very interesting and well-written. Here are some minor comments:

Point 1: I have some concerns about using SG first derivative to correct for baseline. This removes only constant trend. Is it the case for your UV-VIS spectra? There are many better methods suitable for baseline removal, no matter what function it follows. For instance, methods with asymmetric penalised least squares usually perform better. Have you tried any of these?

Response 1:
No. In fact, the authors did not use asymmetric penalised least squares for baseline removal. In this research, we combined three spectral pre-processing of moving averaging smoothing with 5 segments (MAS), standard normal variate (SNV), and Savitzky-Golay first derivative with 11 smoothing gaps and second-order polynomial (SG1d). We have revised this part to better explain how we select the spectral pre-processing.

Since there is no standard protocol for spectral pre-processing, a trial and error approach were adopted. Different spectral pre-processing is available in the Unscrambler X ver. 10.4 (CAMO Software AS, Oslo, Viken, Norway) to reduce or to remove the effect of several different unwanted interfering phenomena such as particle size influence (baseline different and light scattering), etc. As mentioned by Roger et al. (2020) and Bian et al. (2020), it is hard to determine which pre-processing can successfully improve the given original spectral data. For this reason, instead of selecting the best pre-processing, in order to optimize the effect of spectral pre-processing, the combination of several spectral pre-processing was often used. In this study, a selective combination pre-processing strategy was used by combining three different spectral pre-processing of MAS, SNV and SG1d. This combination was done sequentially, e.g. MAS followed by SNV and followed by SG1d.

Savitzky-Golay first derivative with a second-order polynomial and a window size of 11 points (SG1d) was used to cancel the baseline drifts and to enhance small spectral differences (Shawky and Selim, 2019). Due to similarity in coffee species of both samples of wet and dry-processed coffees, it was expected that the spectral difference in peaberry coffee samples due to differences in level of adulteration between wet and dry processed coffees was small. However, at the same time, as a consequence of SG1d derivation, the noises were also enhanced. To avoid this, the spectra were first smoothed using moving averaging smoothing pre-processing as recommended by previous work (Shawky and Selim, 2019). Therefore, in this present study we utilized three sequentially spectral pre-processing: MAS, SNV and SG1d (MAS+SNV+SG1d).

Our approach was previously used by Shawky and Selim (2019), Zhang et al. (2021) and Suhandy and Yulia (2021).

References:

Bian, X.; Wang, K.; Tan, E.; Diwu, P.; Zhang, F.; Guo, Y. A selective ensemble preprocessing strategy for near-infrared spectral quantitative analysis of complex samples. Chemom. Intell. Lab. Syst. 2020, 197, 103916. doi: 10.1016/j.chemolab.2019.103916.

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Zhang, Z.; Wang, Y.; Yan, H.; Chang, X.; Zhou, G.; Zhu, L.; Liu, P.; Guo, S.; Dong, T.T.X.; Duan, J. Rapid geographical origin identification and quality assessment of angelicae sinensis radix by FT-NIR spectroscopy. *J Anal Methods Chem*. 2021, 2021, 1–12. doi: 10.1155/2021/8875876.

Revision in section 2.3 and line 133:

Original sentence:
Since there is no standard protocol for spectral pre-processing, a trial and error approach were adopted. To eliminate noise and systematic spectra variation, three consecutive spectral pre-processing were found to be the best applied: moving averaging with 5 segments (MAS), standard normal variate (SNV), and Savitzky-Golay first derivative with 11 smoothing gaps, and second-order polynomial (SG1d). MAS was widely used to smooth the spectral data before applying various pre-processing [21]. SNV was effective to normalize spectra for cancelling the scattering effect while SG1d was used to correct the baseline effect [22–23].

Revised sentence:
Since there is no standard protocol for spectral pre-processing, a trial and error approach was adopted. Different spectral pre-preprocessing is available in the Unscrambler X ver. 10.4 (CAMO Software AS, Oslo, Viken, Norway) to reduce or to

remove the effect of several different unwanted interfering phenomena such as particle size influence (baseline different and light scattering), etc. As mentioned by Roger et al. [24] and Bian et al. [25], it is hard to determine which pre-processing can successfully improve the given original spectral data. For this reason, instead of selecting the best pre-processing, in order to optimize the effect of spectral pre-processing, the combination of several spectral pre-processing was often used [19]. To eliminate noise and systematic spectra variation, three consecutive spectral pre-processing were found to be the best applied: moving averaging smoothing with 5 segments (MAS), standard normal variate (SNV), and Savitzky-Golay first derivative with 11 smoothing gaps and second-order polynomial (SG1d). MAS was widely used to smooth the spectral data before applying various pre-processing [26]. SNV was effective to normalize spectra for cancelling the scattering effect while SG1d was used to correct the baseline effect [27–28]. Due to similarity in coffee species of both samples of wet and dry-processed coffees, it was expected that the spectral difference in peaberry coffee samples due to differences in level of adulteration between wet and dry processed coffees was small. The SG1d spectral pre-processing was also used to enhance these small spectral differences [26].

The following references have been added in the revised article:

[24] Roger, J.; Biancolillo, A.; Marini, F. Sequential preprocessing through ORThogonalization (SPORT) and its application to near infrared spectroscopy. *Chemom. Intell. Lab. Syst.* 2020, 199, 103975. https://doi:10.1016/j.chemolab.2020.103975.

[25] Bian, X.; Wang, K.; Tan, E.; Diwu, P.; Zhang, F.; Guo, Y. A selective ensemble preprocessing strategy for near-infrared spectral quantitative analysis of complex samples. *Chemom. Intell. Lab. Syst.* 2020, 197, 103916. doi: 10.1016/j.chemolab.2019.103916.

Point 2: Line140: Should be "spectral" instead of "spectra".

Response 2:
Yes. The authors agree to revise this part. The word “spectra” has been replaced by “spectral”.

Original sentence:
PLSR and PCR were developed using spectra data from 250 to 400 nm (number of variables=161).

Revised sentence:
PLSR and PCR were developed using spectral data from 250 to 400 nm (number of variables=161).

Point 3: Line 142: Which of the variables were then finally chosen for PCR? How high loadings were considered, i.e. what threshold was the cut-off?

Response 3: Yes. The authors agree to revise this part.
As it was mentioned in the article, PLSR and PCR were developed using spectral data from 250 to 400 nm (number of variables=161). There is no variable selection applied for both PLSR and PCR method. The variable selection was applied for MLR, since for MLR the number of variables must be less than the number of samples.

For variable selection in MLR, we agree to revise this part to avoid misinterpretation. In MLR, a selected few variables were obtained from a plot of PCA x-loadings. The x-loadings (XLs) in PCA played a crucial role in compressing data, improving modeling efficiency, and reflecting the degree of correlation between several PCs and original variables (Zhao et al., 2021). In this study, the x-loadings of the first 2 PCs were used to identify the important variables.

There is no any threshold as a cut-off value for x-loadings values. Wavelengths that are associated with the positive and negative peaks in the PCA x-loadings plot were used as input. For this reason, based on Figure 4, six wavelengths were used as input variables in MLR: 267, 278, 290, 305, 328 and 345 nm.

Reference:
Zhao, Y.; Fang, S.; Ye, Y.; Yu, K. Chemometric development using portable molecular vibrational spectrometers for rapid evaluation of AVC (*Valsa mali Miyabe* et Yamada) infection of apple trees. *Vib. Spectrosc.* 2021, 114, 103231. doi: 10.1016/j.vibspec.2021.103231.

Revision section 2.3 and line 162:
Original sentences:
In MLR, a selected few variables were obtained from a plot of x-loadings. Wavelengths that are associated with the peaks (both positive and negative) with high x-loadings were used as input.

Revised sentences:
In MLR, a selected few variables were obtained from a plot of PCA x-loadings. Wavelengths that are associated with the positive and negative peaks were used as input.

Revision section 3.2 and line 255:
Original sentences:
It was reported that the caffeine content in dry processing coffees is higher since about 40% of caffeine is removed with pulp during the wet processing [36].

Revised sentence:
It was reported that the caffeine content in dry processing coffees is higher since about 40% of caffeine is removed with pulp during the wet processing [39]. These positive and negative peaks obtained from PCA x-loadings of pre-processed spectral data at 267, 278, 290, 305, 328 and 345 nm were used as input variables for constructing the MLR model.

Point 4: Line 207: Should be Figure 3a.

Response 4: Yes. The authors agree to revise this part. The word "Figure 2 (a)" has been replaced by "Figure 3 (a)".

Original sentence:
Figure 2 (a) shows the scores of the first two principal components (PC1 and PC2) of all coffee samples including wet and dry with 10-50% of corn adulteration using raw spectral data in the range between 250 and 400 nm.

Revised sentence:
Figure 3 (a) shows the scores of the first two principal components (PC1 and PC2) of all coffee samples including wet and dry with 10-50% of corn adulteration using raw spectral data in the range between 250 and 400 nm.

Point 5: How were the data prepared before PCA apart from preprocessing methods?

Response 5:
The spectral data from UV-vis spectrometer was obtained in .csv format. The spectral data preparation before doing PCA and other multivariate analysis including reformat spectral data into .xls instead of .csv and then import the .xls spectral data into the Unscrambler X ver. 10.4 (CAMO Software AS, Oslo, Viken, Norway).

Revision section 2.2 line 127:

Original sentences:
Raw UV spectral data was obtained for aqueous samples in the range between 250 and 400 nm with 1 nm interval using a UV-visible spectrometer (Genesys™ 10S UV-Vis, Thermo Scientific, Waltham, MA, USA).

Revised sentences:
Raw UV-Vis spectral data was obtained for aqueous samples in the range between 250 and 400 nm with 1 nm interval using a UV-visible spectrometer (Genesys™ 10S UV-Vis, Thermo Scientific, Waltham, MA, USA) in .csv format. After reformatting into .xls, the raw spectral data were imported to the Unscrambler X ver. 10.4 (CAMO Software AS, Oslo, Viken, Norway) for chemometrics analysis.

Point 6: Line 213: Should be Figure 3b.

Response 6:
Yes. The authors agree to revise this part. The word "Figure 2 (b)" has been replaced by "Figure 3 (b)".

Original sentence:
A better PCA score plot was achieved using pre-processed spectral data in the range between 250 and 400 nm as presented in Figure 2 (b).

Revised sentence:
A better PCA score plot was achieved using pre-processed spectral data in the range between 250 and 400 nm as presented in Figure 3 (b).

Point 7: Line 215: Should be Figure 4.

Response 7:
Yes. The authors agree to revise this part. The word "Figure 3" has been replaced by "Figure 4".

Original sentence:
Figure 3 shows the loadings plot of PC1 and PC2 using pre-processed spectral data.

Revised sentence:
Figure 4 shows the loadings plot of PC1 and PC2 using pre-processed spectral data.

Point 8: Line 251: Should be Table 3.

Response 8:
Yes. The authors agree to revise this part. The word "Table 2" has been replaced by "Table 3".

Original sentence:
The results are presented in Table 2.

Revised sentence:
The results are presented in Table 3.

Point 9: Line 292: Should be Table 4.

Response 9:

Yes. The authors agree to revise this part. The word “Table 3” has been replaced by “Table 4”.

Original sentence:
The results are presented in Table 3.

Revised sentence:
The results are presented in Table 4.

Response to Reviewer 2 Comments

This manuscript reported corn adulteration in peaberry coffees by UV-Vis spectroscopy. This work is helpful to establish a reliable, low-cost method in coffee adulteration method. However, the overall scientific value of this work remains incomplete. Some of the issues should be resolved.

Point 1: This work achieved quantification as their primary method towards adulteration. However, classification using PCA scores and PLS-DA should be evaluated when detecting fraud. Based on Figure 5 and Table 4, the overall PLSR for quantification still yields high errors in the test sets, which may be the shortcoming as UV-Vis is low-cost and imprecise. However, again, it should be justified the actual classification between pure/adulterated coffee at the claimed LOD around 20% is significant. Otherwise, this work provides few useful information for practical application.

Response 1:

The main advantages of UV-vis spectroscopy are spectrometer in this region is relatively low cost and it is available to most standard laboratories, a green technology without chemical waste during sample extraction and simple in sample preparation. However, a multivariate analysis is an obligation as a tandem for UV-Vis spectroscopy due to large spectral data usually necessary to develop reliable results and various overlaps in the spectral data. Recently, UV-Vis spectroscopy has been well reported for the quantification of food fraud including coffee and honey (Suhandy and Yulia, 2021; Valinger et al., 2021).

The objective of this current research is to evaluate a spectroscopic method based on UV spectroscopy and chemometrics to quantify the corn adulteration in coffee involving two common types of bean processing of wet and dry-processed methods. Using the same spectroscopic system, both qualitative and quantitative studies on peaberry and civet or luwak coffee have been reported with the acceptable result (Suhandy and Yulia, 2017a; Suhandy and Yulia, 2017b). UV-Vis spectral information to be used for robust and simple discrimination of ground pure peaberry and ground normal coffee samples from the same bean processing method. The two supervised discrimination methods investigated, SIMCA and PLS-DA, provided satisfactory classification rates, 100% for accuracy, sensitivity, and specificity (Suhandy and Yulia, 2017a). Using the developed PLS regression model, a prediction for quantification of luwak content was calculated and resulted in satisfactory prediction performance

with high both RPD and RER values (RPD=6.11 and RER=17.86) in the prediction step (Suhandy and Yulia, 2017b).

In this study, we justified the acceptance of the developed multivariate model by using two parameters of RPD and RER. In UV spectroscopy, RPD and RER are also used in the previous study to evaluate the acceptance of the developed model (Valinger et al. 2021). According to Chang et al. (2001) and Valinger et al. (2021) models with RPD<1.4 are considered non-reliable for prediction; models considered as representative are for 1.4<RPD<2.0 and models with the RPD>2.0 are excellent for description and prediction of experimental data. According to Jia et al. (2017), RER less than 3 indicated that practical utility was little, from 3 to 10 indicated that the model was good practical utility, and RER more than 10 indicated excellent accuracy. Based on Table 4, the prediction result for samples with the same bean processing method is acceptable for corn quantification with RPD more than 2 and RER more than 7.

For example, in Table 4, the individual wet PLSR model was used to predict corn adulteration in three types of prediction samples: wet, dry, and combined prediction samples. The model is working well only for wet prediction samples (both calibration and prediction samples are wet-processed peaberry coffee). The RPD is 3.96 and the RER is 11.20. A similar result was obtained for the individual dry PLSR model. The RPD of 3.33 and RER of 9.43 could be obtained. The individual dry PLSR model also failed to quantify corn adulteration in wet and combined prediction samples. However, our proposed method to develop a global PLSR model is promising. The global PLSR model worked better with RPD more than 2 for wet, dry, and combined prediction samples. The RER is also close to 7 (in the range of 3 to 10) for all prediction samples. Using this consideration, our current research is important to show that postharvest treatments such as bean processing highly affected the robustness of the developed UV-Vis model for quantification of corn adulteration in peaberry coffee with different bean processing methods. Our current research also successfully demonstrated a promising method of using the global PLSR model which can handle the effect of the different bean processing method in the developed PLSR model. This global PLSR model may be useful for practical application to quantify corn adulteration in peaberry coffee incorporated with different bean processing methods.

References:

Chang, C.-W.; Laird, D.A.; Mausbach, M. J.; Hurburgh, C.H. Near-Infrared reflectance spectroscopy–principal components regression analyses of soil properties. *Soil Sci. Soc. Am. J.* 2001, 65, 480–490. doi: 10.2136/sssaj2001.652480x.

Jia, B.; Yoon, S.-C.; Zhuang, H.; Wang, W.; Li, C. Prediction of pH of fresh chicken breast fillets by VNIR hyperspectral imaging. *J. Food Eng*. 2017, 208, 57–65. doi: 10.1016/j.jfoodeng.2017.03.023.

Suhandy, D.; Yulia, M. Peaberry coffee discrimination using UV-visible spectroscopy combined with SIMCA and PLS-DA. *Int. J. Food Prop*. 2017a, 20, S331–S339. doi: 10.1080/10942912.2017.1296861.

Suhandy, D.; Yulia, M. The use of partial least square regression and spectral data in uv-visible region for quantification of adulteration in Indonesian palm civet coffee. *Int. J. Food. Sci.* 2017b, 2017, 6274178. doi: 10.1155/2017/6274178.

Suhandy, D.; Yulia, M. The use of ultraviolet (uv) spectroscopy and chemometrics to quantify the percentages of adulteration in Kalosi ground roasted specialty coffee. *J. Eng. Sci. Technol*. 2021, 16, 350–364.

Valinger, D.; Longin, L.; Grbes, F.; Benkovic, M.; Jurina, T.; Kljusuric, J.G.; Tusek, A.J. Detection of honey adulteration – The potential of UV-VIS and NIR spectroscopy coupled with multivariate analysis. *LWT*. 2021, 145, 111316. doi: 10.1016/j.lwt.2021.111316.

Revision in section 3.4 line 345:

Original sentence:
The RPD_P was higher than 2 for both individual predictions of wet and dry samples and combined samples.

Revised sentences:
The RPD_P was higher than 2 for both individual predictions of wet and dry samples and combined samples. According to Chang et al. [43] and Valinger et al. [44] models with RPD >2.0 are excellent description and prediction of experimental data.

Two references have been added in the revised article:

[43] Chang, C.-W.; Laird, D.A.; Mausbach, M. J.; Hurburgh, C.H. Near-Infrared reflectance spectroscopy–principal components regression analyses of soil properties. *Soil Sci. Soc. Am. J*. 2001, 65, 480–490. doi: 10.2136/sssaj2001.652480x.

[44] Valinger, D.; Longin, L.; Grbes, F.; Benkovic, M.; Jurina, T.; Kljusuric, J.G.; Tusek, A.J. Detection of honey adulteration – The potential of UV-VIS and NIR spectroscopy coupled with multivariate analysis. *LWT*. 2021, 145, 111316. doi: 10.1016/j.lwt.2021.111316.

The calculation of LOD and LOQ obtained in this study should be revised. According to Milani et al. (2020) and Rambla-Alegre et al. (2012), the LOD and LOQ should be calculated based on calibration curve, not prediction curve. For this reason, we revised Figure 5 and recalculate the LOD and LOQ. The obtained LOD and LOQ using UV-Vis spectroscopy in this study were in the range of 12.48%~16.84% and 41.61%~56.14%. In this present study, a calibration and validation regression model were developed using corn adulterated samples in the range of 10-50% (w/w). However, the obtained LOD and LOQ in this study suggested we extend the range of corn adulteration up to more than 50%. For this reason, it needs an improvement for practical application. However, in Indonesia, adulteration more than 50% of specialty coffee is commonly found for economically motivated adulteration (Suhandy and Yulia, 2021).

Revision in section 3.3 and 3.4

Figure 5 and its explanation has been replaced from section 3.4 to section 3.3 in the revised article.

Figure 5 has been revised.

Original sentences:
Figure 5 shows plots of prediction for individual and combined prediction samples predicted using the global PLSR model. Visually, it can be noticed that the residuals of prediction were randomly scattered closely to the regression line (bias is close to 0) for individual and combined prediction samples. The SEP and slope for individual prediction wet samples were 6.35% (w/w) and 0.91 resulting in the LOD and LOQ of 20.93% (w/w) and 69.78% (w/w), respectively. Similarly, the LOD and LOQ for individual prediction dry samples were 23.15% (w/w) and 77.18% (w/w). For combined prediction samples, the LOD and LOQ were 21.63% (w/w) and 72.10% (w/w). Comparing to previous works, our result was inferior. For example, Milani et al. [15] reported satisfactory LOD values of 0.31-0.86% using the NMR spectroscopy with a different roasting profile. Sezer eta l. [16] reported a quantitative approach using LIBS for coffee adulteration with different adulterants (corn, wheat, and chickpea) and resulted in a promising result with the LOD below 0.6% could be obtained. The obtained LOD and LOQ using UV spectroscopy in this study were in the range of 20.93%~23.15% and 69.78%~72.10%. For this reason, it needs an improvement for practical application. However, in Indonesia, adulteration more than 50% of specialty coffee is commonly found for economically motivated adulteration [12].

Revised sentences:
Figure 5 shows plots of the best PLSR calibration model for individual wet, dry and combined calibration samples. Visually, it can be noticed that the residuals of

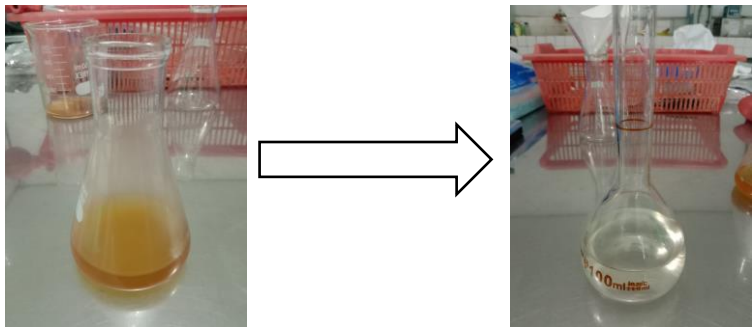
calibration were randomly scattered closely to the regression line (bias is close to 0) for individual and combined calibration samples. The SEC and slope for individual calibration wet samples were 3.87% (w/w) and 0.93 resulting in the LOD and LOQ of 12.48% (w/w) and 41.61% (w/w), respectively. Similarly, the LOD and LOQ for individual calibration dry samples were 12.88% (w/w) and 42.93% (w/w). For combined calibration samples, the LOD and LOQ were 16.84% (w/w) and 56.14% (w/w). Comparing to previous works, our result was inferior. For example, Milani et al. [15] reported satisfactory LOD values of 0.31-0.86% using the NMR spectroscopy with a different roasting profile. Sezer eta l. [16] reported a quantitative approach using LIBS for coffee adulteration with different adulterants (corn, wheat, and chickpea) and resulted in a promising result with the LOD below 0.6% could be obtained. The obtained LOD and LOQ using UV-Vis spectroscopy in this study were in the range of 12.48%~16.84% and 41.61%~56.14%. In this present study, a calibration and validation regression model were developed using corn adulterated samples in the range of 10-50% (w/w). However, the obtained LOD and LOQ in this study suggested we extend the range of corn adulteration up to more than 50%. For this reason, it needs an improvement for practical application. However, in Indonesia, adulteration more than 50% of specialty coffee is commonly found for economically motivated adulteration [12].

Point 2: Although this work focused on UV frequency range, most UV spectroscopy also extends to the Visible region. Additionally, the visual appearance for adulteration is different enough in own observation. The author should refer UV as UV-Vis all along the manuscript. Also, the full UV-vis range should be inspected for modelling, at least solid reasons should be given why the visible range is not considered, along with the data.

Response 2:
Yes. The authors agree to revise this part.
Gad et al. (2013) analyzed UV spectroscopy in the range 200-400 nm for quality control of thyme. In contrast, Souto et al. (2015) utilized UV-Vis spectra in the range of 239-405 nm. Souto et al. (2010) performed a UV-Vis spectral measurement in the range of 225-353 nm with a 1 nm interval. In this present research, we acquired spectral data in the range between 250 and 400 nm with a 1 nm interval. For this reason, the authors agree to refer UV as UV-Vis all along with the manuscript.

The authors agree that the visual appearance for adulteration is different enough in own observation as seen in Figure 1 in this current research. The adulterated peaberry wet-processed samples are darker than that of peaberry dry ones. However, it is visually difficult to discriminate between the different levels of adulteration in both wet and dry-processed adulterated peaberry samples. In this UV-Vis spectroscopy, the spectral acquisition was done in aqueous samples. After extraction protocol

including dilution with hot distilled water, all samples were similar in color as we can see in the following Figure I.



Extracted coffee aqueous samples Diluted coffee aqueous samples

Figure I. Visual appearance of peaberry coffee samples after extraction and dilution.

In this present study, we used a UV-Vis spectrometer with the possible range for spectral acquisition from 190 to 1100 nm (default range). In previous research, using the same system, the authors reported peaberry and civet or luwak coffee authentication with the full spectral acquisition from 190 to 700 nm (from UV to full visible light region) (Suhandy and Yulia, 2017a; Suhandy and Yulia, 2017b). The typical feature of original or raw UV spectral data is highly noisy with very high absorbance (more than 2) especially in the interval of 190-250 nm (high-frequency noise). This raw spectral data is rich in unrelated information such as background information and systematic noise coming from the influences of light scattering, differences in path length, sample particle size, and other factors. Similar results were reported for UV spectral data of ground roasted coffee from Brazil (Souto et al., 2010) and ground roasted coffee from Indonesia (Suhandy and Yulia, 2017a; Suhandy and Yulia, 2017b) with absorbance intensity of more than 2. Dankowska et al. (2017) also reported UV-Vis absorption spectra of aqueous extracts of the genuine Arabica and Robusta coffee samples and their mixtures in the range 190–700 nm. Diniz et al. (2016) obtained absorbance spectra of the simple tea infusions in the range of 190-800 nm with a very high absorbance of more than 2 was observed in the range of 190-240 nm. The typical raw UV-Vis spectra also have very low absorbance intensity in visible light (from 400 to 700 nm). The final model for constructing qualitative and quantitative analysis was developed using a selected wavelength instead of using a full spectrum. For example, Suhandy and Yulia (2017a) utilized spectral data in the range of 190-400 nm instead of using the full region 190-700 nm. Suhandy and Yulia (2017b) used original and pre-processing spectra in the range 200-450 nm for the determination of luwak content using PLS regression. Based on these previously reported studies, in this present research, we acquire UV-Vis spectral data from 250 to 400 nm. In our opinion, this selected region acquisition is more effective with rich in information and faster in spectral acquisition comparing to that of full-spectrum acquisition

Herewith the authors would like to show visually several reported UV-Vis spectral data showing a relatively high absorbance (more than 2) in the interval of 190-250 nm and very low absorbance (close to zero) in the interval of 400-700 nm.

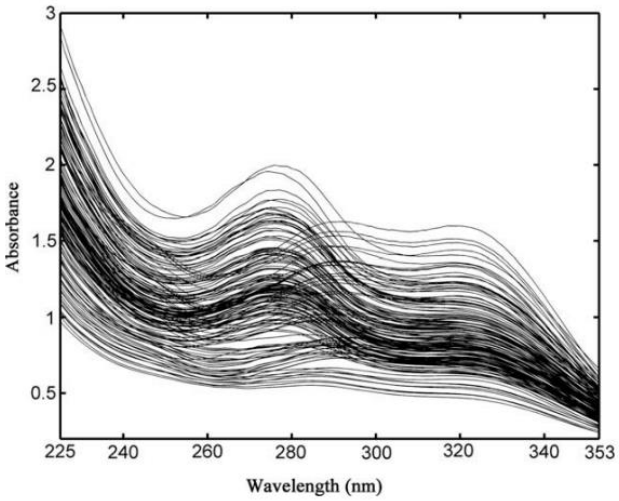


Fig. 1. UV–Vis spectra of the 175 coffee samples.

UV-Vis spectral data from Souto et al. (2010).

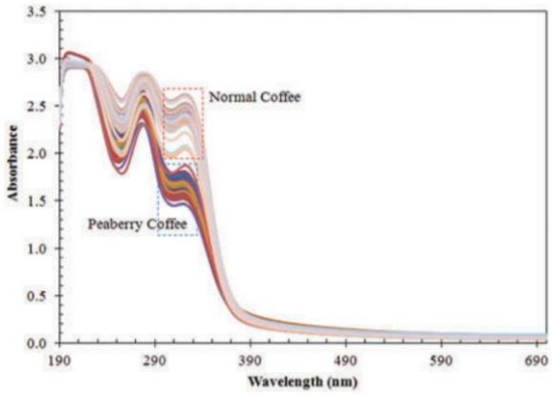


Figure 1. Original spectra of peaberry and normal coffee samples in ultraviolet-visible region (190–700 nm).

UV-Vis spectral data from Suhandy and Yulia (2017a).

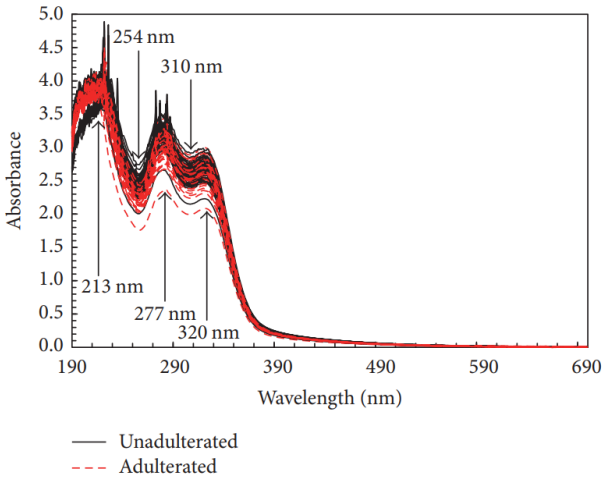


FIGURE 1: Original spectra of unadulterated and adulterated coffee samples in the UV-Vis region.

UV-Vis spectral data from Suhandy and Yulia (2017b).

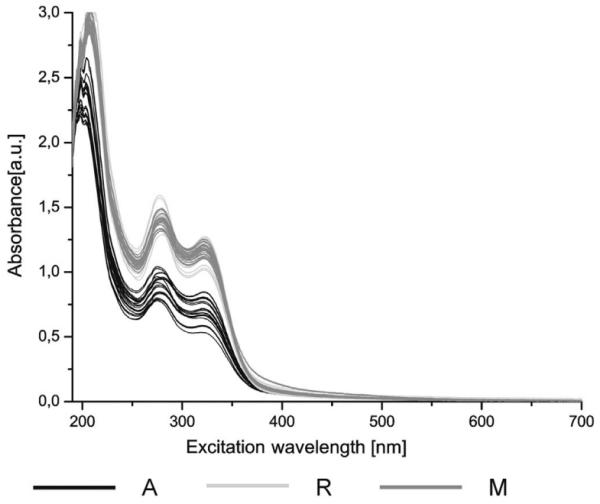


Fig. 2. UV-Vis spectra of *Coffea arabica*, *Coffea robusta*, and their mixtures (diluted 1:120 v/v in water) (A - *Coffea arabica*, R - *Coffea robusta*, M - mixtures of *Coffea arabica* and *Coffea robusta*).

UV-Vis spectral data from Dankowska et al. (2017).

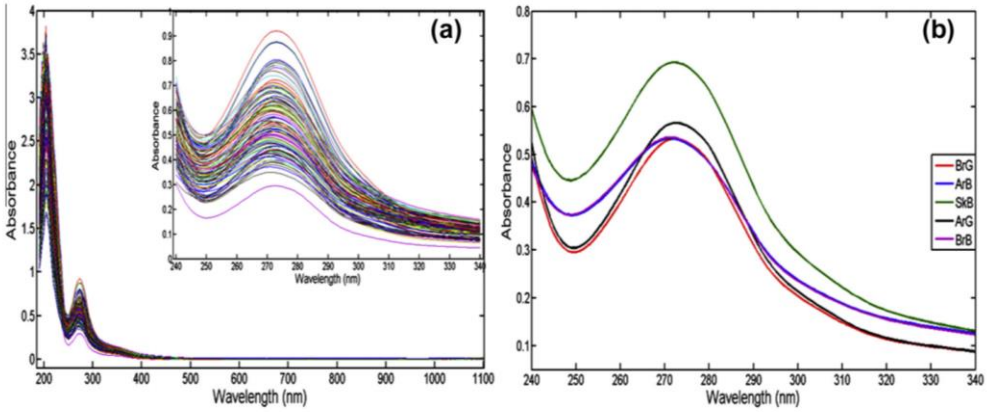


Fig. 1. (a) Raw UV–Vis spectra of all studied tea samples. (b) Mean spectra of the five studied tea classes. Argentinean green (ArG, ▬), Brazilian green (BrG, ▬), Argentinean black (ArB, ▬), Brazilian black (BrB, ▬), and Sri Lankan black (SkB, ▬).

UV-vis spectral data of tea samples from Diniz et al. (2016).

References:

Dankowska, A.; Domagała, A.; Kowalewski, W. Quantification of coffea arabica and coffea canephora var. robusta concentration in blends by means of synchronous fluorescence and uv-vis spectroscopies. *Talanta* 2017, 172, 215–220. doi: 10.1016/j.talanta.2017.05.036.

Diniz, P.H.G.D.; Barbosa, M.F.; de Melo Milanez, K.D.T.; Pistonesi, M.F.; de Araújo, M.C.U. Using uv–vis spectroscopy for simultaneous geographical and varietal classification of tea infusions simulating a home-made tea cup. *Food Chem*. 2016, 192, 374–379. doi: 10.1016/j.foodchem.2015.07.022.

Gad, H.A.; El-Ahmady, S.H.; Abou-Shoer, M.I.; Al-Azizi, M.M. (2013). A modern approach to the authentication and quality assessment of thyme using uv spectroscopy and chemometric analysis. *Phytochem. Anal.* 2013, 24(6), 520–526. doi: 10.1002/pca.2426.

Souto, U.T.C.P.; Barbosa, M.F.; Dantas, H.V.; de Pontes, A.S.; Lyra, W.S.; Diniz, P.H.G.D.; de Araújo, M.C.U.; da Silva, E.C. Identification of adulteration in ground roasted coffees using UV–Vis spectroscopy and SPA-LDA. *LWT - Food Sci. Technol.* 2015, 63(2), 1037–1041. doi: 10.1016/j.lwt.2015.04.003.

Souto, U.T.C.P.; Pontes, M.J.C.; Silva, E.C.; Galvão, R.K.H.; Araújo, M.C.U.; Sanches, F.A.C.; Cunha, F.A.S.; Oliveira, M.S.R. UV–Vis spectrometric classification of coffees by SPA–LDA. *Food Chem*. 2010, 119(1), 368–371. doi: 10.1016/j.foodchem.2009.05.078.

Suhandy, D.; Yulia, M. Peaberry coffee discrimination using uv-visible spectroscopy combined with SIMCA and PLS-DA. *Int. J. Food Prop*. 2017a, 20(sup1), S331–S339. doi: 10.1080/10942912.2017.1296861.

Suhandy, D.; Yulia, M. The use of partial least square regression and spectral data in uv-visible region for quantification of adulteration in Indonesian palm civet coffee. *Int. J. Food Sci*. 2017b, 2017, 1–7. doi: 10.1155/2017/6274178.

Response to Reviewer 3 Comments

The present manuscript presents a method for the quantification of corn adulteration in peaberry ground roasted coffee that have been wet or dry processed. The work uses multivariate linear regression methods to model and predict the amount of adulteration, and PLS has proved to be the best method for both wet and dry processed samples.

The manuscript is very well written and is very clear throughout the text. However, even though the premise is very interesting (to use a more affordable approach, such as UV spectroscopy, for the detection of the adulteration), it carries some flaws that I believe are critical to the acceptance of the results. Hence, I do not recommend its publication in the current form. In my opinion, the main points are:

Point 1: Major flaws in interpreting the spectra and the loadings information throughout the text. As described in section 2.3., the data was pre-processed by smoothing, standardization, baseline correction and differentiation. The last method is, indeed, much used for regression. The first derivative of a spectrum will show how it varies, where the peaks are presented as zero (since it is the local maxima), the regions where the original spectrum signal are increasing are presented as positive values and the regions where the original spectrum signal are decreasing are presented as negative values. Figure 2(a) and 2(b) clearly shows that the pre-processed data presents the first derivative. The "peaks" in the pre-processed spectra (270, 290, 315 and 345 nm) are not peaks in the raw spectra. Interpretations were made in sections 3.1. and 3.2., where these values were treated, discussed, and compared to the literature. This should be closely looked by the authors, so no misinterpretation remains. If comparisons should be made, it should take into consideration that the data represents the first derivative.

Response 1: The authors agree to revise this part.

In this research, we combined 3 spectral pre-processing of moving averaging smoothing with 5 segments (MAS), standard normal variate (SNV), and Savitzky-Golay first derivative with 11 smoothing gaps and second-order polynomial (SG1d).

The typical feature of original or raw UV-Vis spectral data obtained in several previous studies is highly noisy with very high absorbance (more than 2) especially in the interval of 190-250 nm (high-frequency noise). That is why in this current study we started the spectral acquisition from 250 nm. Another feature of UV spectral data is a broad spectrum with highly correlated and overlapped data. For this reason, spectral pre-processing is necessary before applying further analysis of PCA or PLS

regression as demonstrated by several previous works (Suhandy and Yulia, 2021a; Suhandy and Yulia, 2021b). In this study as we can see in Figure 2 (a) that the raw UV-Vis spectra are broad and overlap, hence it is hard to differentiate between wet and dry-processed adulterated peaberry. However, in Figure 2 (a) several broad peaks could be identified around 260, 275, 305, and 325 nm. In Figure 2(b) the pre-processed spectral data is looked better with clear peaks and no overlapping between wet and dry-processed adulterated peaberry coffees. The authors agree that the peaks we obtained in the pre-processed spectral data (270, 290, 315, and 345 nm) are not peaks in the raw UV-Vis spectra. For this reason, the authors agree that the comparison of the peaks obtained in this study with previously published works should be clarified at first derivative spectral data. Therefore, the authors agree to revise the discussion in section 3.1.

References:

Suhandy, D.; Yulia, M. The use of ultraviolet (uv) spectroscopy and chemometrics to quantify the percentages of adulteration in Kalosi ground roasted specialty coffee. *J. Eng. Sci. Technol.* 2021a, 16, 350–364.

Suhandy, D.; Yulia, M. Classification of Lampung robusta specialty coffee according to differences in cherry processing methods using uv spectroscopy and chemometrics. *Agric.* 2021b, 11(2), 109. doi: 10.3390/agriculture11020109.

Revision in section 3.1 line 212:

Original sentences:
Several positive and negative peaks were observed clearly in the pre-processed spectral data. The highest positive peak at 270 nm was closely related to the C=O chromophore in caffeine molecules as reported by some previous works [30–31], indicating the significant difference of the caffeine content in adulterated wet and dry peaberry coffees. The negative peaks at 290 and 345 nm was corresponding with the absorbance of chlorogenic acids (CGA) [31]. Navarra et al. [32] reported a wavelength at 330 nm for the CGA absorbance when ethanol was used as the solvent. Dankowska et al. [31] reported wavelength at 320 nm as one of the negative peaks found in the spectral data of arabica and robusta coffee and its adulteration using water as solvent. In this study, with water used as the solvent, the peak of CGA was shifted to the longer wavelength at 345 nm. This shifting phenomenon was supported by the previous work by Souto et al. [30], with water used as the solvent, they found wavelength shifting of CGA from 320 nm to 325 nm.

Revised sentences:
Several positive and negative peaks were observed clearly in the pre-processed spectral data (MAS+SNV+SG1d). The highest positive peak at 270 nm of pre-processed

spectra was closely related to the C=O chromophore in caffeine molecules as reported by some previous works [35–36], indicating the significant difference of the caffeine content in adulterated wet and dry peaberry coffees. The negative peaks at 290 and 345 nm of pre-processed spectra was corresponding with the absorbance of chlorogenic acids (CGA) of raw UV-Vis spectra in previous work [36]. Navarra et al. [37] reported a wavelength at 330 nm for the CGA absorbance when ethanol was used as the solvent. Dankowska et al. [36] reported wavelength at 320 nm as one of the negative peaks found in the raw UV-Vis spectral data of arabica and robusta coffee and its adulteration using water as solvent. In this study, with water used as the solvent, the peak of CGA of pre-processed spectral data was shifted to the longer wavelength at 345 nm. This shifting phenomenon was also found by the previous work by Souto et al. [35], with water used as the solvent, they found wavelength shifting of CGA from 320 nm to 325 nm in raw UV-Vis spectral data.

Section 3.2 is about PCA and its plots. There are two PCA plots in section 3.2: score and x-loadings plot. The x-loadings (XLs) in PCA played a crucial role in compressing data, improving modeling efficiency, and reflecting the degree of correlation between several PCs and original variables (Zhao et al., 2021). In this study, the x-loadings of the first 2 PCs were used to identify the important variables. Based on Figure 4, six wavelengths were identified as important variables: 267, 278, 290, 305, 328, and 345 nm. These wavelengths have a significant contribution to the separation between the adulterated peaberry wet and dry coffees. In this section, we discussed the possible reason why those wavelengths are important. However, to avoid misinterpretation the authors agree to revise this part.

Revision section 3.2 line 250:

Original sentences:
Souto et al. [30] reported the maxima electronic absorption of trigonelline at 275 nm, caffeine at 280 nm, and caffeic acid at 325 nm. However, the adulterated peaberry dry coffees were mainly discriminated by the negative peak for PC1 at the wavelength of 278 nm, indicating that the adulterated peaberry dry samples coffees contain high contents of caffeine. It was supported by previous work [34]. It was reported that the caffeine content in dry processing coffees is higher since about 40% of caffeine is removed with pulp during the wet processing [34].

Revised sentences:
Souto et al. [35] reported the maxima electronic absorption of trigonelline at 275 nm, caffeine at 280 nm, and caffeic acid at 325 nm using raw UV-Vis spectra. However, the adulterated peaberry dry coffees were mainly discriminated by the negative peak for PC1 at the wavelength of 278 nm, indicating that the adulterated peaberry dry samples coffees contain high contents of caffeine. It was supported by previous work

[39]. It was reported that the caffeine content in dry processing coffees is higher since about 40% of caffeine is removed with pulp during the wet processing [39]. These positive and negative peaks obtained from PCA x-loadings of pre-processed spectral data at 267, 278, 290, 305, 328, and 345 nm were used as input variables for constructing the MLR model.

Point 2: Why was PCA only conducted with the adulterated samples? Was it important to separate the wet from dry sample or to separate the pure from adulterated sample? I believe that the latter is the most important one. As I will discuss in the next point, calibration was not good because the variability between each replicate sample was very high. Take Figure 5 as an example. For solutions with the same amount of adulteration, the model did not see them as equal, but as quite different. Perhaps lack of reproduction is related to the variability of the beans? Is it related to any of the process conducted? The fact is that the solutions that should represent the same thing does not. That is what the regression error is so big. Not because it is not good, or invalid, but because it is trying to model data that are not good. Perhaps the objective of the work shouldn't be to quantify (because it is not predicting anything, since the LOQ is even higher than the range studied), but to classify the samples using multivariate methods. If the "pure" sample data were added to the PCA, maybe PCs will be able to separate them from the adulterated ones. This could be a tool to detect if it is the real peaberry coffee beans or not. PCA could even be refined to try to separate the samples in groups (e.g., based on the amount of corn bean).

Response 2:

The authors agree that both issues are important. Several previous works reported both qualitative and quantitative studies on ground roasted coffee adulteration (Suhandy and Yulia, 2021a; Suhandy and Yulia 2021b). Qualitatively, it is important to discriminate between pure and adulterated coffees (Suhandy and Yulia, 2021a). Using the same system, Suhandy and Yulia (20121a) reported the possible application of UV spectroscopy and chemometrics method for ground roasted Kalosi coffee authentication both in qualitative (classify the samples into a low, middle, and a high degree of adulteration) and quantitative studies (quantify the percentage of adulteration in Kalosi ground roasted coffee) with acceptable results.

Based on our previous work using the same spectroscopic system (as seen below), UV-Vis spectroscopy with PCA can separate between the pure and adulterated coffee samples.

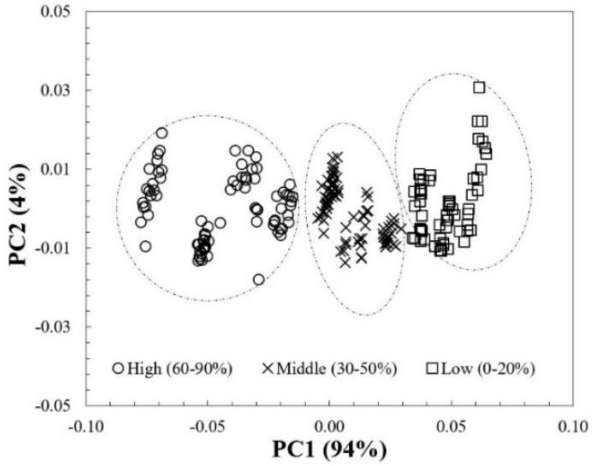


Fig. 6. PCA score plot of Kalosi coffee samples with different percentages of adulteration calculated using pre-processed spectra in the range of 200-400 nm.

UV-Vis spectroscopy and PCA could be effectively discriminate the pure (0% adulteration) from its adulterated Kalosi arabica coffee samples (Suhandy and Yulia, 2021a).

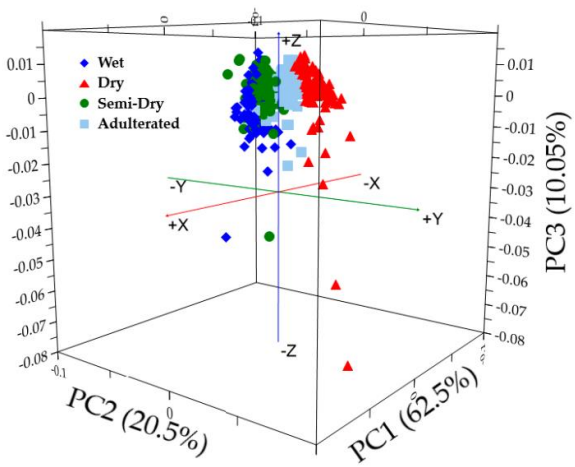


Figure 3. Scores plot of PC1, PC2 and PC3 from PCA analysis in the interval of 230–350 nm of combined pre-treated spectral data.

UV-Vis spectroscopy and PCA were used to separate between the pure wet, dry, semi-dry, and adulterated Lampung robusta coffee (Suhandy and Yulia, 2021b).

However, in the aforementioned studies, no reported works included the influence of other important postharvest factors especially the bean processing method in the developed calibration models. Therefore, in this present research, we evaluated a spectroscopic method based on UV-Vis spectroscopy and chemometrics to quantify the corn adulteration in coffee involving two common types of bean processing of wet and dry-processed methods. From this point, the authors decide to keep the

originality of the present research to investigate a robust calibration model using three different linear regression methods including PLSR, MLR, and PCR for quantification of the corn adulteration in peaberry specialty coffee incorporated with different bean processing methods. For this reason, in this current research, the use of PCA to separate the wet from dry-processed peaberry sample is more important.

Figure 5 has been revised. The authors agree that one possible reason for the high regression error in Figure 5 is the influence of different bean processing methods (high variability of the beans). Figure 5 is the result of the calibration using the best wet, dry and global PLSR model. The higher regression error was identified for the global PLSR calibration model. The global PLSR model was developed using combined samples from wet and dry-processed peaberry coffees. As it can be seen in Table 4, the RPD and RER are becoming lower indicating the influence of different bean processing methods on prediction performance is significant. This is one important finding of this present research. Additionally, the proposed global PLSR model used in this study can improve the prediction performance (with acceptable RPD and RER values) indicating another important finding of this analytical method to handle the influence of different bean processing methods.

References:

Suhandy, D.; Yulia, M. The use of ultraviolet (uv) spectroscopy and chemometrics to quantify the percentages of adulteration in Kalosi ground roasted specialty coffee. *J. Eng. Sci. Technol.* 2021a, 16, 350–364.

Suhandy, D.; Yulia, M. Classification of Lampung robusta specialty coffee according to differences in cherry processing methods using uv spectroscopy and chemometrics. *Agric.* 2021b, 11(2), 109. doi: 10.3390/agriculture11020109.

Point 3: A The regression is not quantifying anything! As I mentioned in the point above, it is very difficult to accept a model in which the LOQ is substantially higher than the maximum concentration value used for the construction of the model. Your consideration that it needs a future improvement would be reasonable, but only if your own model could be validated. The results show that you would only be able to quantify adulterations over 70%. However, you never used any solutions above this concentration. Your model for quantification was constructed entirely below the limit that it says that it can quantify. When you compare your model to other methods in the literature that present such small LOD, it seems that, even though the UV spectroscopy is more accessible, it is not worth to substitute using another technique such as NMR or LIBS.

Response 3: Yes. The authors agree to revise this part.

At first, in this study, we justified the acceptance of the developed regression model by using two parameters of RPD and RER. In UV-Vis spectroscopy and other spectroscopic methods, RPD and RER are also frequently used in the previous study to evaluate the acceptance of the developed calibration model (Valinger et al. 2021). According to Chang et al. (2001) and Valinger et al. (2021) models with RPD<1.4 are considered non-reliable for prediction; models considered as representative are for 1.4<RPD<2.0 and models with the RPD>2.0 are excellent for description and prediction of experimental data. According to Jia et al. (2017), RER less than 3 indicated that practical utility was little, from 3 to 10 indicated that the model was good practical utility, and RER more than 10 indicated excellent accuracy. Based on Table 4, the prediction result for samples with the same bean processing method is acceptable for corn quantification with RPD more than 2 and RER more than 7.

For example, in Table 4, the individual wet PLSR model was used to predict corn adulteration in three types of prediction samples: wet, dry, and combined prediction samples. The model is working well only for wet prediction samples (both calibration and prediction samples are wet-processed peaberry coffee). The RPD is 3.96 and the RER is 11.20. A similar result was obtained for the individual dry PLSR model. The RPD of 3.33 and RER of 9.43 could be obtained. The individual dry PLSR model also failed to quantify corn adulteration in wet and combined prediction samples. However, our proposed method to develop a global PLSR model is promising. The global PLSR model worked better with RPD more than 2 for wet, dry, and combined prediction samples. The RER is also close to 7 (in the range of 3 to 10) for all prediction samples. Using this consideration, our current research is important to show that postharvest treatments such as bean processing highly affected the robustness of the developed UV-Vis model for quantification of corn adulteration in peaberry coffee with different bean processing methods. Our current research also successfully demonstrated a promising method of using the global PLSR model which can handle the effect of the different bean processing methods in the developed PLSR model. This global PLSR model with improvement may be useful for practical application to quantify corn adulteration in peaberry coffee incorporated with different bean processing methods.

The calculation of LOD and LOQ obtained in this study should be revised. According to Milani et al. (2020) and Rambla-Alegre et al. (2012), the LOD and LOQ should be calculated based on calibration curve, not prediction curve. For this reason, we revised Figure 5 and recalculate the LOD and LOQ. The obtained LOD and LOQ using UV-Vis spectroscopy in this study were in the range of 12.48%~16.84% and 41.61%~56.14%. In this present study, a calibration and validation regression model were developed using corn adulterated samples in the range of 10-50% (w/w). However, the obtained LOD and LOQ in this study suggested we extend the range of corn adulteration up to more than 50%. For this reason, it needs an improvement for practical application. However, in Indonesia, adulteration more than 50% of specialty

coffee is commonly found for economically motivated adulteration (Suhandy and Yulia, 2021).

References:

Chang, C.-W.; Laird, D.A.; Mausbach, M. J.; Hurburgh, C.H. Near-Infrared reflectance spectroscopy–principal components regression analyses of soil properties. *Soil Sci. Soc. Am. J.* 2001, 65, 480–490. doi: 10.2136/sssaj2001.652480x.

Jia, B.; Yoon, S.-C.; Zhuang, H.; Wang, W.; Li, C. Prediction of pH of fresh chicken breast fillets by VNIR hyperspectral imaging. *J. Food Eng.* 2017, 208, 57–65. doi: 10.1016/j.jfoodeng.2017.03.023.

Milani, M.I.; Rossini, E.L.; Catelani, T.A.; Pezza, L.; Toci, A.T.; Pezza, H.R. Authentication of roasted and ground coffee samples containing multiple adulterants using NMR and a chemometric approach. *Food Control*, 2020, 112, 107104. doi: 10.1016/j.foodcont.2020.107104.

Rambla-Alegre, M.; Esteve-Romero, J.; Carda-Broch, S. Is it really necessary to validate an analytical method or not? That is the question. *J. Chromatogr. A*, 2012, 1232, 101–109. doi: 10.1016/j.chroma.2011.10.050.

Suhandy, D.; Yulia, M. The use of ultraviolet (uv) spectroscopy and chemometrics to quantify the percentages of adulteration in Kalosi ground roasted specialty coffee. *J. Eng. Sci. Technol.* 2021, 16, 350–364.

Valinger, D.; Longin, L.; Grbes, F.; Benkovic, M.; Jurina, T.; Kljusuric, J.G.; Tusek, A.J. Detection of honey adulteration – The potential of UV-VIS and NIR spectroscopy coupled with multivariate analysis. *LWT*. 2021, 145, 111316. doi: 10.1016/j.lwt.2021.111316.

Revision in section 3.4 line 345:

Original sentence:
The RPD_P was higher than 2 for both individual predictions of wet and dry samples and combined samples.

Revised sentences:
The RPD_P was higher than 2 for both individual predictions of wet and dry samples and combined samples. According to Chang et al. [43] and Valinger et al. [44] models with RPD >2.0 are excellent descriptions and predictions of experimental data.

Two references have been added in the revised article:

[43] Chang, C.-W.; Laird, D.A.; Mausbach, M. J.; Hurburgh, C.H. Near-Infrared reflectance spectroscopy–principal components regression analyses of soil properties. *Soil Sci. Soc. Am. J.* 2001, 65, 480–490. doi: 10.2136/sssaj2001.652480x.

[44] Valinger, D.; Longin, L.; Grbes, F.; Benkovic, M.; Jurina, T.; Kljusuric, J.G.; Tusek, A.J. Detection of honey adulteration – The potential of UV-VIS and NIR spectroscopy coupled with multivariate analysis. *LWT.* 2021, 145, 111316. doi: 10.1016/j.lwt.2021.111316.

Revision in section 3.3 and 3.4

Figure 5 and its explanation has been replaced from section 3.4 to section 3.3 in the revised article.

Figure 5 has been revised.

Original sentences:
Figure 5 shows plots of prediction for individual and combined prediction samples predicted using the global PLSR model. Visually, it can be noticed that the residuals of prediction were randomly scattered closely to the regression line (bias is close to 0) for individual and combined prediction samples. The SEP and slope for individual prediction wet samples were 6.35% (w/w) and 0.91 resulting in the LOD and LOQ of 20.93% (w/w) and 69.78% (w/w), respectively. Similarly, the LOD and LOQ for individual prediction dry samples were 23.15% (w/w) and 77.18% (w/w). For combined prediction samples, the LOD and LOQ were 21.63% (w/w) and 72.10% (w/w). Comparing to previous works, our result was inferior. For example, Milani et al. [15] reported satisfactory LOD values of 0.31-0.86% using the NMR spectroscopy with a different roasting profile. Sezer eta l. [16] reported a quantitative approach using LIBS for coffee adulteration with different adulterants (corn, wheat, and chickpea) and resulted in a promising result with the LOD below 0.6% could be obtained. The obtained LOD and LOQ using UV spectroscopy in this study were in the range of 20.93%~23.15% and 69.78%~72.10%. For this reason, it needs an improvement for practical application. However, in Indonesia, adulteration more than 50% of specialty coffee is commonly found for economically motivated adulteration [12].

Revised sentences:
Figure 5 shows plots of the best PLSR calibration model for individual wet, dry and combined calibration samples. Visually, it can be noticed that the residuals of calibration were randomly scattered closely to the regression line (bias is close to 0) for individual and combined calibration samples. The SEC and slope for individual

calibration wet samples were 3.87% (w/w) and 0.93 resulting in the LOD and LOQ of 12.48% (w/w) and 41.61% (w/w), respectively. Similarly, the LOD and LOQ for individual calibration dry samples were 12.88% (w/w) and 42.93% (w/w). For combined calibration samples, the LOD and LOQ were 16.84% (w/w) and 56.14% (w/w). Comparing to previous works, our result was inferior. For example, Milani et al. [15] reported satisfactory LOD values of 0.31-0.86% using the NMR spectroscopy with a different roasting profile. Sezer eta l. [16] reported a quantitative approach using LIBS for coffee adulteration with different adulterants (corn, wheat, and chickpea) and resulted in a promising result with the LOD below 0.6% could be obtained. The obtained LOD and LOQ using UV-Vis spectroscopy in this study were in the range of 12.48%~16.84% and 41.61%~56.14%. In this present study, a calibration and validation regression model were developed using corn adulterated samples in the range of 10-50% (w/w). However, the obtained LOD and LOQ in this study suggested we extend the range of corn adulteration up to more than 50%. For this reason, it needs an improvement for practical application. However, in Indonesia, adulteration more than 50% of specialty coffee is commonly found for economically motivated adulteration [12].

Revision in section conclusion:

Original sentence:
The proposed UV spectroscopy and global PLSR model detected an admixture of corn in the peaberry ground roasted coffee in the range 10% to 50% with the LOD values of 20.93-23.15% cloud be reported for individual and combined samples

Revised sentences:
The proposed UV-Vis spectroscopy and global PLSR model detected an admixture of corn in the peaberry ground roasted coffee in the range 10% to 50% with the LOD values of 12.48%~16.84% could be reported for individual and combined samples.

Table has been made on Table 2. LOD and LOQ moved to calibration steps.

Point 4: The abstract says that R^2 for PLSR is "more than 0.70", but in fact is way more than that.

Response 4: Yes. The authors agree to revise this part.

In the calibration, the R^2 of the best PLSR model for individual wet, dry and combined samples were 0.93, 0.92, and 0.88, respectively. In the validation step, the R^2 of the best PLSR model for individual wet, dry and combined samples were 0.89, 0.89, and 0.83, respectively. Therefore, the range of R^2 for the best PLSR model is 0.83~0.93.

Revision in section abstract line 19:

Original sentence:
The best calibration model for individual wet and dry and combined samples were obtained for the PLSR model with a coefficient of determination more than 0.70 and RMSE below 6% (w/w) for both calibration and validation.

Revised sentence:
The best calibration model for individual wet and dry and combined samples were obtained for the PLSR model with a coefficient of determination in the range of 0.83~0.93 and RMSE below 6% (w/w) for calibration and validation.

Point 5: Introduction can be improved to give a background to the reader regarding the corn beans as adulterants. You only cite that "the adulteration is frequently happened in the form of ground roasted coffee". How important is the adulteration with corn beans? Is it relevant?

Response 5: The authors agree to revise this part. More explanation has been added in the introduction. The reason why using corn for diluents in coffee adulteration has been described.

Coffee adulteration may be performed by changing the quality of beans or adding other low-cost coffee and non-coffee materials as described by previously reported studies: robusta coffee (Garrett et al., 2012), inferior quality of arabica coffee (Toledo et al., 2014), mixed of four materials (coffee husks, spent coffee ground, barley, and corn) (Reis et al., 2016), wheat, corn, and chickpea (Sezer et al., 2018), soybeans, green mung beans and spent coffee grounds (Cheah and Fang, 2020), and coffee husks, soybean, corn, barley, rice, and wheat (Milani et al., 2020). In this study, corn was selected as an adulterant material due to its low cost and huge availability in the Indonesian market. For this reason, in a real situation, the adulteration of ground roasted peaberry coffee involved the intentional addition of finely ground corn. Additionally, corn is one of the most used diluents in coffee adulteration as reported in several previous works (Reis et al., 2016; Sezer et al., 2018; Milani et al., 2020; Ferreira et al., 2021; Pereira et al., 2021).

References:

Cheah, W.L.; Fang, M. HPLC-based chemometric analysis for coffee adulteration. *Foods*, 2020, 9(7), 880. doi: 10.3390/foods9070880.

Ferreira, T.; Galluzzi, L.; de Paulis, T.; Farah, A. Three centuries on the science of coffee authenticity control. *Food Res. Int.* 2021, 149, 110690. doi: 10.1016/j.foodres.2021.110690.

Garrett, R.; Vaz, B.G.; Hovell, A.M.C.; Eberlin, M.N.; Rezende, C.M. Arabica and robusta coffees: identification of major polar compounds and quantification of blends by direct-infusion electrospray ionization–mass spectrometry. *J. Agric. Food Chem*. 2012, 60(17), 4253–4258. doi: 10.1021/jf300388m.

Milani, M.I.; Rossini, E.L.; Catelani, T.A.; Pezza, L.; Toci, A.T.; Pezza, H.R. Authentication of roasted and ground coffee samples containing multiple adulterants using NMR and a chemometric approach. *Food Control*, 2020, 112, 107104. doi: 10.1016/j.foodcont.2020.107104.

Pereira, L.H.; Catelani, T.A.; Costa, E.D.M.; Garcia, J.S.; Trevisan, M.G. Coffee adulterant quantification by derivative thermogravimetry and chemometrics analysis. *J. Therm. Anal. Calorim*. 2021. doi: 10.1007/s10973-021-11016-6.

Reis, N.; Franca, A.S.; Oliveira, L.S. Concomitant use of Fourier transforms infrared attenuated total reflectance spectroscopy and chemometrics for quantification of multiple adulterants in roasted and ground coffee. *J. Spectrosc.* 2016, 2016, 4974173. doi: 10.1155/2016/4974173.

Sezer, B.; Apaydin, H.; Bilge, G.; Boyaci, I.H. Coffee arabica adulteration: Detection of wheat, corn and chickpea. *Food Chem.* 2018, 264, 142–148. doi: 10.1016/j.foodchem.2018.05.037.

Toledo, B.R.; Hantao, L.W.; Ho, T.D.; Augusto, F.; Anderson, J.L. A chemometric approach toward the detection and quantification of coffee adulteration by solid-phase microextraction using polymeric ionic liquid sorbent coatings. *J. Chromatogr. A*. 2014, 1346, 1–7. doi: 10.1016/j.chroma.2014.04.035.

Revision in section 1. Introduction and line 78:

Original sentence:
Therefore, in this present research, we evaluated a spectroscopic method based on UV spectroscopy and chemometrics to quantify the corn adulteration in coffee involving two common types of bean processing of wet and dry processed methods.

Revised sentences:
In this study, corn was selected as an adulterant material due to its low cost and huge availability in the Indonesian market. Additionally, corn is one of the most used diluents in coffee adulteration as reported in several previous works [15-16, 21-23]. Therefore, in this present research, we evaluated a spectroscopic method based on UV-Vis spectroscopy and chemometrics to quantify the corn adulteration in coffee involving two common types of bean processing of wet and dry-processed methods.

The following references have been added to the revised article:

[21] Reis, N.; Franca, A.S.; Oliveira, L.S. Concomitant use of Fourier transforms infrared attenuated total reflectance spectroscopy and chemometrics for quantification of multiple adulterants in roasted and ground coffee. *J. Spectrosc.* 2016, 2016, 4974173. doi: 10.1155/2016/4974173.

[22] Ferreira, T.; Galluzzi, L.; de Paulis, T.; Farah, A. Three centuries on the science of coffee authenticity control. *Food Res. Int.* 2021, 149, 110690. doi: 10.1016/j.foodres.2021.110690.

[23] Pereira, L.H.; Catelani, T.A.; Costa, E.D.M.; Garcia, J.S.; Trevisan, M.G. Coffee adulterant quantification by derivative thermogravimetry and chemometrics analysis. *J. Therm. Anal. Calorim*. 2021. doi: 10.1007/s10973-021-11016-6.

Point 6: The method is not clear regarding how you achieve 100 and 99 samples, when you describe that it was prepared only the 10%, 20%, 30%, 40% and 50% concentrations. The reader can only guess that all are replicates when looking at Figure 5. Why were this procedure chosen? Why not use other concentrations?

Response 6: Yes. The authors agree to revise this part. More explanation has been added in the revised article.

In this research, the wet and dry-processed peaberry ground roasted coffees were intentionally adulterated with the ground roasted corn in the range of 10-50% (w/w) with an increment of 10% (w/w). This adulteration range was chosen according to several previous works [15–16]. It is also the most common adulteration level found in the Indonesian markets [12]. Therefore, there are five levels of corn adulteration for wet and dry-processed peaberry coffee. Total 199 samples (1 gram each) of adulterated peaberry dry and wet-processed coffees were provided. It consists of 20 samples for each level of corn adulteration resulted in a total of 100 samples for dry-processed peaberry coffees and 99 samples for wet-processed peaberry coffees (19 samples were provided at a level of 40% for wet-processed peaberry coffees).

Why an increment of 10% was used? the increment percentage of adulteration used by previous studies in coffee adulteration is varied as presented in Table I. For example, the coffee blends in increment of 2.5% (v/v) for calibration and increment of 2% (v/v) for validation were provided for quantitative coffee adulteration using the LIBS method (Sezer et al., 2018). Milani et al. (2020) selected adulterant percentages in increments of 1.5, 2.5, 5, 15, and 25% (w/w) for authentication of ground roasted coffee samples with multiple adulterants using NMR spectroscopy and chemometric. Cheah and Fang (2020) prepared coffee adulterated samples in increments of 5%, 10%, and

20% (w/w) for detecting coffee adulteration using HPLC-based chemometric analysis. Suhandy and Yulia (2021) prepared a wide range of adulteration, the ratio between ground roasted Kalosi coffee and coffee skins are 0 to 90% (w/w) in increment of 10% from low (0-20% w/w), middle (30-50% w/w) and a high degree of adulteration (60-90% w/w) for calibration, validation, and prediction. Nunez et al. (2021) used a 20% of increment of coffee adulteration in a calibration set for the detection and quantitation of adulterated coffee samples by chemometrics and HPLC-FLD fingerprinting.

Table I. Range of adulteration percentages and their increment used in ground roasted coffee authentication.

References	Range of adulteration percentages (%)	
	Calibration set	Prediction set
Sezer et al. (2018)	2.5-60% (v/v) in increment of 2.5%	2-50% (v/v) in increment of 2%
Milani et al. (2020)	1.0, 2.5, 5.0, 10, 25, and 50% (w/w)	1.0, 2.5, 5.0, 10, 25, and 50% (w/w)
Cheah and Fang (2020)	5, 10, 20,40, and 60% (w/w)	5, 10, 20,40, and 60% (w/w)
Suhandy and Yulia (2021)	0 to 90% (w/w) in increment of 10%	0 to 90% (w/w) in increment of 10%
Nunez et al. (2021)	20, 40, 60 and 80%	15, 25, 50, 75 and 85%

References:

Cheah, W.L.; Fang, M. HPLC-based chemometric analysis for coffee adulteration. *Foods*, 2020, 9, 880. doi: 10.3390/foods9070880.

Milani, M.I.; Rossini, E.L.; Catelani, T.A.; Pezza, L.; Toci, A.T.; Pezza, H.R. Authentication of roasted and ground coffee samples containing multiple adulterants using NMR and a chemometric approach. *Food Control*, 2020, 112, 107104. doi: 10.1016/j.foodcont.2020.107104.

Nunez, N.; Saurina, J.; Nunez, O. Non-targeted HPLC-FLD fingerprinting for the detection and quantitation of adulterated coffee samples by chemometrics. *Food Control*, 2021, 124, 107912. doi: 10.1016/j.foodcont.2021.10791.

Sezer, B.; Apaydin, H.; Bilge, G.; Boyaci, I.H. (2018). *Coffee arabica* adulteration: detection of wheat, corn and chickpea. *Food Chem*. 2018, 264, 142–148. doi: 10.1016/j.foodchem.2018.05.037.

Suhandy, D.; Yulia, M. The use of ultraviolet (uv) spectroscopy and chemometrics to quantify the percentages of adulteration in Kalosi ground roasted specialty coffee. *J. Eng. Sci. Technol.* 2021, 16, 350–364.

Revision in section 2.1 and line 113:

Original sentences:
Total 100 and 99 samples (1 gram each) of adulterated peaberry wet and dry-processed coffees were provided.

Revised sentences:
Total 199 samples (1 gram each) of adulterated peaberry dry and wet-processed coffees were provided. It consists of 20 samples for each level of corn adulteration resulted in a total of 100 samples for dry-processed peaberry coffees and 99 samples for wet-processed peaberry coffees (19 samples were provided at a level of 40% for wet-processed peaberry coffees).

Point 7: Page 3, line 124 says that 3 pre-processing methods were used, but 4 are described.

Response 7:
In this research, we combined 3 spectral pre-processing of moving averaging smoothing with 5 segments (MAS), standard normal variate (SNV), and Savitzky-Golay first derivative with 11 smoothing gaps and second-order polynomial (SG1d). We have revised this part to better explain how we select the spectral pre-processing.

Since there is no standard protocol for spectral pre-processing, a trial and error approach was adopted. Different spectral pre-processing is available in the Unscrambler X ver. 10.4 (CAMO Software AS, Oslo, Viken, Norway) to reduce or to remove the effect of several different unwanted interfering phenomena such as particle size influence (baseline different and light scattering), etc. As mentioned by Roger et al. (2020) and Bian et al. (2020), it is hard to determine which pre-processing can successfully improve the given original spectral data. For this reason, instead of selecting the best pre-processing, to optimize the effect of spectral pre-processing, the combination of several spectral pre-processing was often used. In this study, a selective combination pre-processing strategy was used by combining three different pre-processing of MAS, SNV, and SG1d. This combination was done sequentially, e.g. MAS followed by SNV and followed by SG1d.

Savitzky-Golay first derivative with a second-order polynomial and a window size of 11 points (SG1d) was used to cancel the baseline drifts and to enhance small spectral differences (Shawky and Selim, 2019). Due to similarity in coffee species of both samples of wet and dry-processed coffees, it was expected that the spectral difference

in peaberry coffee samples due to differences in the level of adulteration between wet and dry-processed coffees was small. However, at the same time, as a consequence of SG1d derivation, the noises were also enhanced. To avoid this, the spectra were first smoothed using moving averaging smoothing pre-processing as recommended by previous work (Shawky and Selim, 2019). Therefore, in this present study, we utilized three sequentially spectral pre-processing: MAS, SNV, and SG1d (MAS+SNV+SG1d). Our approach was previously used by Shawky and Selim (2019) and Zhang et al. (2021).

References:

Bian, X.; Wang, K.; Tan, E.; Diwu, P.; Zhang, F.; Guo, Y. A selective ensemble preprocessing strategy for near-infrared spectral quantitative analysis of complex samples. *Chemom. Intell. Lab. Syst.* 2020, 197, 103916. doi: 10.1016/j.chemolab.2019.103916.

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Revision section 2.3 and line 133:

Original sentence:
Since there is no standard protocol for spectral pre-processing, a trial and error approach were adopted. To eliminate noise and systematic spectra variation, three consecutive spectral pre-processing were found to be the best applied: moving averaging with 5 segments (MAS), standard normal variate (SNV), and Savitzky-Golay first derivative with 11 smoothing gaps, and second-order polynomial (SG1d). MAS was widely used to smooth the spectral data before applying various pre-processing [21]. SNV was effective to normalize spectra for cancelling the scattering effect while SG1d was used to correct the baseline effect [22–23].

Revised sentence:

Since there is no standard protocol for spectral pre-processing, a trial and error approach was adopted. Different spectral pre-preprocessing is available in the Un-scrambler X ver. 10.4 (CAMO Software AS, Oslo, Viken, Norway) to reduce or to remove the effect of several different unwanted interfering phenomena such as particle size influence (baseline different and light scattering), etc. As mentioned by Roger et al. [24] and Bian et al. [25], it is hard to determine which pre-processing can successfully improve the given original spectral data. For this reason, instead of selecting the best pre-processing, to optimize the effect of spectral pre-processing, the combination of several spectral pre-processing was often used. To eliminate noise and systematic spectra variation, three consecutive spectral pre-processing were found to be the best applied: moving averaging smoothing with 5 segments (MAS), standard normal variate (SNV), and Savitzky-Golay first derivative with 11 smoothing gaps and second-order polynomial (SG1d). MAS was widely used to smooth the spectral data before applying various pre-processing [26]. SNV was effective to normalize spectra for canceling the scattering effect while SG1d was used to correct the baseline effect [27–28]. Due to similarity in coffee species of both samples of wet and dry-processed coffees, it was expected that the spectral difference in peaberry coffee samples due to differences in the level of adulteration between wet and dry-processed coffees was small. The SG1d spectral pre-processing was also used to enhance these small spectral differences [26].

Point 8: Page 4, lines 148 and 149 says that three samples were selected for the calibration set out of 5. From what is show in Table 1, this is probably mistaken. I believe it should say that 4 out of 5 was chosen as the calibration set.

Response 8: Yes. The authors agree to revise this part. Yes, it should be 4 out of 5 that were chosen as the calibration set.

Revision section 2.3 and line 168

Original sentence:
The procedure of this separation of the samples was as follows: order the samples concerning the corn adulteration level (from minimum to maximum values), then three samples were selected every five samples for the calibration and the rest for prediction.

Revised sentence:
The procedure of this separation of the samples was as follows: order the samples concerning the corn adulteration level (from minimum to maximum values), then four samples were selected every five samples for the calibration and the rest for prediction.

Point 9: Page 8, lines 270-282 are unnecessary in my opinion. Comparing the behavior of adulterations in honey, measured by ion mobility, shouldn't be the same as in the system studied in this work.

Response 9: Yes. The authors agree to revise this part.
We remove these sentences in the revised article. Reference of Aliano-Gonzalez et al. (2020) also has been removed.

Point 10: It is not clear to what variables were used for the MLR model. Which frequency was chosen for that? Information should be provided.

Response 10: Yes. The authors agree to revise this part.
For variable selection in MLR, we agree to revise this part to avoid misinterpretation. In MLR, a selected few variables were obtained from a plot of x-loadings. There is no threshold as a cut-off value for x-loadings values. Wavelengths that are associated with the positive and negative peaks were used as input. For this reason, based on Figure 4, six wavelengths were used as input variables in MLR: 267, 278, 290, 305, 328, and 345 nm.

Revision in section 2.3 and line 162:
Original sentences:
In MLR, a selected few variables were obtained from a plot of x-loadings. Wavelengths that are associated with the peaks (both positive and negative) with high x-loadings were used as input.

Revised sentences:
In MLR, a selected few variables were obtained from a plot of x-loadings. Wavelengths that are associated with the positive and negative peaks were used as input.

Revision in section 3.2 and line 255:

Original sentences:
It was reported that the caffeine content in dry processing coffees is higher since about 40% of caffeine is removed with pulp during the wet processing [34].

Revised sentence:
It was reported that the caffeine content in dry processing coffees is higher since about 40% of caffeine is removed with pulp during the wet processing [39]. These positive and negative peaks obtained from PCA x-loadings of pre-processed spectral data at 267, 278, 290, 305, 328, and 345 nm were used as input variables for constructing the MLR model.